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OPTIMIZATION OF PT-DOPED KOCITE(R) (TRADEMARK) ELECTRODES IN H3--ETC(U)
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UOP, Incorporated
Des Plaines, Illinois

October 1976

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OPTIMIZATION OF PT-DOPED KOCITE[R] ELECTRODES IN H_3PO_4 FUEL CELLS

by

L. B. Welsh, R. W. Leyerle

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The use of UOP Pt-impregnated Kocite materials as low cost air and/or fuel electrocatalysts in phosphoric acid electrolyte fuel cells is being optimized. Kocite[R] materials are composite structures consisting of pyropolymers chemically bonded to refractory substrates. Fuel cell electrodes are fabricated from these materials and tested as anodes or cathodes in model fuel cells with Teflon bonded Pt black counter electrodes.

Kocite is being produced in 200-400 gram quantities from a high surface area

alumina with a particle size of ∿500 nm. From the Pt-impregnated Kocite, electrodes with a porous graphite paper current collector are being fabricated by both hand and machine calendering operations with 0.125, 0.25, and 0.50 mg/cm^2 Pt loadings. When tested in model phosphoric acid fuel cells with 2 mg/cm^2 Pt black counter electrodes, full-cell IR-free performances of 0.62 and 0.71 volts as air and oxygen cathodes, respectively, have been obtained at 100 ma/cm^2 and 149°C using 0.25 mg/cm^2 Pt-loaded Kocite electrodes. Lifetime testing of Kocite electrodes as cathodes indicates no catalyst deterioration occurs for periods in excess of 500 hours for temperatures from 140 to 180°C.

In order to optimize the structure of Kocite electrodes, the teflon content, density, and thickness of the Kocite catalyst layer have been systematically varied. Testing results obtained to date indicate relatively high porosity catalyst layers with ∿30 wt.% teflon and a 2.5 mg/cm^2 or smaller Kocite loading will achieve the best performance levels consistent with the required resistance to electrolyte flooding.

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I. INTRODUCTION

1.1 Objectives

The objective of this program is to optimize the performance of low-cost air and fuel electrodes fabricated from platinum-impregnated proprietary UOP Kocite[R] materials for use in phosphoric acid electrolyte fuel cells within the temperature range of 140-180°C. In order to accomplish this objective, various Kocite formulations are produced and fabricated into state-of-the-art fuel cell electrodes. These electrodes are tested as anodes or cathodes in model fuel cells with conventional platinum black electrodes as counter electrodes. Long term (500 hour) endurance testing is completed on promising electrodes.

This report summarizes the results obtained during the second five months of this program. The fuel cell performance levels achieved during this period using platinum-doped Kocite[R] fuel cell electrodes will be discussed, along with the research plans for the duration of the contract.

1.2 Background

The current research program follows an evaluation of the performance of Kocite[R] materials in fuel cell electrodes for phosphoric acid electrolyte fuel cells[(1,2)] completed under USA/MERADCOM contract number DAAK02-75-C-0011. The program organization established under this previous contract[(1,2)] has been continued. This involved a joint

[1] L. B. Welsh, G. L. Hervert, D. H. Spielberg, K. J. Youtsey, "Carbonaceous Catalysts for H_3PO_4 Fuel Cells", MERDC Contract DAAK02-75-C-0011, Interim Technical Report, March, 1975.

[2] L. B. Welsh, R. W. Leyerle, G. L. Hervert, and K. J. Youtsey, "Carbonaceous Catalysts for H_3PO_4 Fuel Cells", MERDC Contract DAAK02-72-C-0011, Final Technical Report, September, 1975.

program with UOP Inc. as the prime contractor and Energy Research Corporation (a subsidiary of St. Joe Minerals Corporation) as subcontractor. A broad division of effort is made on the basis of the UOP capability to produce Pt-impregnated Kocite^R materials and the ERC capability to fabricate and optimize electrode structures. Testing and diagnostic evaluation of these electrodes are carried out jointly by UOP and ERC.

1.3 Review of Previous Results

1.3.1 Kocite^R Materials

Kocite^R materials are composite structures consisting of a thin film, semiconducting pyropolymer chemically bonded to the surface of an electrically insulating, high surface area, refractory substrate. The materials used to produce Kocite^R can vary widely but commonly include gamma alumina as the refractory substrate and hydrocarbons such as cyclohexane and benzene as the pyrolyzable material or pyropolymer precursor. Pyrolysis temperatures are commonly in the range of 400 to 900°C. The electrical conductivity of Kocite materials can be varied widely in a controlled manner by adjusting one or more of the following parameters: (a) substrate composition and surface area, (b) pyrolyzable material, and (c) reaction conditions (e.g., reaction time, temperature, ambient gases). The electrochemical behavior may also depend on these parameters. The application of Kocite materials consists of their utilization as high surface area extenders for catalytically active materials (transition metals, etc.). The preparation of Kocite materials and their analysis have been discussed in earlier reports.[1,2]

1.3.2 Results Obtained During the First Five Month Period

In this section the results obtained during the first five month period of the present contract will be briefly summarized.(3)

The main effort during this period was directed toward producing large Kocite^R batches from small particle size alumina bases ($\leq 10^3$ nm) and then impregnating these materials at platinum loadings up to 15 wt.% Pt. Some of the resulting electrocatalysts were evaluated in electrodes. With the use of these smaller particle size alumina bases, a substantial improvement in both the performance levels and oxygen gain were obtained(3) relative to the best performance levels achieved under the previous contract,(1,2) DAAK02-75-C-0011. In addition, electrode testing at 160 and 180°C was expanded substantially. Some of the most important achievements of this period are listed below.

(A) Kocite batches of several hundred grams were prepared in a rotary kiln from both ground Kaiser Medium Alumina ($\sim 1.2 \times 10^3$ nm dia.) and Alcoa Hydral 705 ($\sim$500 nm dia.) for fabrication into machine calendered electrodes. The pore volume distribution of Kocites made from these base materials was measured and found to be substantially different, suggesting both the wetting properties and gas diffusion losses may differ in electrodes fabricated from these materials.

(B) Substantial electrode performance testing was accomplished with both 0.25 and 0.50 mg/cm^2 Pt loaded Kocite electrodes used as cathodes. At 100 ma/cm^2 and 140°C, fuel cell IR free performance levels of 0.55 and 0.66 volts were obtained with 0.25 mg/cm^2

[3] L. B. Welsh, R. W. Leyerle, G. L. Hervert, "Optimization of Pt-Doped Kocite^R Electrodes in H_3PO_4 Fuel Cells", MERADCOM Contract DAAG53-76-C-0014, Interim Technical Report, March, 1976.

Pt-loaded Kocite electrodes operated on air and oxygen, respectively. With 0.50 mg/cm^2 Pt-loaded Kocite electrodes, performance levels of 0.63 and 0.74 were obtained on air and oxygen, respectively, under the same operating conditions.

(C) Kocite electrodes were shown to give stable performance as air cathodes at 100 ma/cm^2 for periods in excess of 500 hours at temperatures from 140 to 180°C.

1.4 Program of the Second Five Month Period

The main effort during this period has been to produce a large number of electrode batches of differing structural parameters from a single Kocite electrocatalyst batch. In order to optimize the structure of Kocite electrodes, the electrode structure parameters have been varied systematically with respect to catalyst-layer teflon content, density, and thickness. For better uniformity and reproducibility, these electrodes are now made by machine calendering rather than hand rolling. Although the variation of electrode structure parameters is incomplete, a substantial amount of test data on machine calendered electrodes has been obtained which indicates how Kocite electrodes may be optimized. In addition, several electrocatalysts made from the $\sim 1.2 \times 10^3$ nm diameter Kaiser Medium Alumina and the ~ 500 nm diameter Hydral 705 have been fabricated into electrodes and tested. The work performed during the second five months of the current contract can be divided into the following areas.

(A) Several large batches (200 to 400 gms) of Kocite have been prepared from both ground Kaiser Medium Alumina base ($\sim 1.2 \times 10^3$ nm dia.) and Alcoa Hydral 705 ($\sim$500 nm dia.) using benzene as the pyropolymer precursor. These batches have been sent to ERC for fabrication into machine calendered electrodes.

(B) Several large batches of Kocite have been prepared in a rotary kiln reactor using different pyropolymer precursors (decane, cyclohexane, pyridine) to determine the optimal production parameters of these Kocites for fuel cell applications.

(C) Techniques for the Pt-impregnation of the Hydral 705 alumina based Kocite have been optimized.

(D) Using a large Hydral-based Kocite batch, the effects of catalyst-layer Teflon content, thickness, and density on the cathode performance of Kocite electrodes between 140 and 180°C are being determined. This batch has been impregnated at the 5 wt.% Pt level.

(E) Substantial performance and lifetime test data have been accumulated on electrodes fabricated from a variety of Pt-impregnated Kocite electrocatalysts. These data include limited data on the performance of Kocite electrodes as anodes at 180°C. During this period, fuel cell hardware and components have been standardized.

(F) Preliminary data have been obtained on the degree of Pt particle growth during electrode fabrication.

A detailed discussion of the results obtained in each of these areas is presented in the following sections.

II. PT-DOPED KOCITE[R] ELECTROCATALYSTS

The preparation and physical properties of the platinum-doped Kocite batches, from which electrodes have been fabricated, are discussed in this section. Some of these properties are listed in Table I. As the general preparation and platinum impregnation techniques have been discussed in earlier reports(1-3), only specific alternations of these techniques will be discussed here.

Three different alumina base materials were used to prepare the electrocatalysts listed in Table I. The 3173-173B, 3173-173C, and 3289-25F batches were prepared from Kaiser Medium Alumina ground to a mean particle size of $\sim 2.2 \times 10^3$ nm, while batch 3380-104B is made using the same alumina ground to a mean particle size of $\sim 1.2 \times 10^3$ nm. The other four batches were made from the Hydral 705 alumina with a mean particle size of 500 nm. As discussed in the last report(3), the pore volume distributions of these electrocatalysts are quite different. The Kaiser based materials have a broad, two peaked distribution with peaks near 4.0 and 8.0 nm, while the Hydral based materials have a narrow distribution with a single peak near 4.0 nm. From the results reported earlier(1-3), both the pore volume distribution and mean particle size are expected to affect the Kocite electrode performance.

With the same surface area and resistivity the carbon content of a Hydral based Kocite is substantially smaller than that of a Kaiser based Kocite. Typical carbon contents are about 35 wt.% for the Kaiser based Kocites and 24 wt.% for the Hydral based Kocites.

The characteristics of Kocite batches 3173-173B and 3173-173C have been included in Table I although no new results have been obtained with these electrocatalysts during the second five month period. The results obtained

Table I

Characteristics of Pt-impregnated Kocite[R] Preparations

Batch Number	3173-173B	3173-173C	3289-25F	3380-31B	3380-31E	3380-31K	3380-104B	3380-125B
Analysis, Wt.%								
Pt	1.98	4.78	9.835	4.91	11.78	5.0	4.72	4.85
C	34.7	33.20	36.80	24.09	24.09	24.09	37.3	19.08
Physical Characteristics								
ρ @ 25°C, Ω-cm	0.017	0.014	0.051	0.036	0.036	0.036	0.032	0.13
Apparent Bulk Density, g/ml	0.343	0.375	0.30	0.28	0.28	0.28	0.398	0.32
Surface Area, M/g	69	81	70	69	69	69	79	80
Pore Volume, ml/g	0.18	0.19	0.18	0.09	0.09	0.09	0.17	0.11
Average Pore Diameter, nm	10.4	9.4	10.3	5.2	5.2	5.2	8.6	5.5
Kocite[R] Substrate								
Type	γ-Al_2O_3	γ-Al_2O_3	γ-Al_2O_3	$\gamma+\chi$-Al_2O_3	$\gamma+\chi$-Al_2O_3	$\gamma+\chi$-Al_2O_3	γ-Al_2O_3	$\gamma+\chi$-Al_2O_3
Size Range, 10^3 nm	2.2	2.2	2.2	0.5	0.5	0.5	1.2	0.5
Pyrolysis Temperature (°C)	900	900	900	850	850	850	860	800
Pyropolymer Precursor	C_6H_6	C_6H_6	C_6H_6	C_6H_6	C_6H_6	C_6H_6	C_6H_6	C_6H_6
Pt Addition	To Product	To Product	To Product	To Product	To Product	To Product	To Product	To Product
Pt Reduction Temperature (°C)	260	260	260	260	260	260	260	260

with these materials provide baseline data on which to judge improvements in performance resulting from the various steps taken to optimize Kocite electrocatalysts.

The platinum impregnation of Kocite materials has been described in earlier reports.[1-3] In order to determine if each electrocatalyst has been prepared with a high platinum dispersion, an X-ray linewidth analysis of the platinum crystallite size was performed. The results of this analysis are given in Table II for the electrocatalysts listed in Table I. The percentage of Pt particles having diameters between 25 and 100 nm can be determined, as well as the average diameter of the Pt particles in this size range, when the X-ray data are compared against those of a standard. The numbers listed under agglomeration give the fraction of Pt atoms in Pt particles with a diameter greater than $\sim$2.5 nm. The numbers are only relative because particles smaller than 2.5 nm diameter are not measured as contributing to the Pt X-ray linewidth. The agglomeration numbers are normalized as discussed in an earlier report. With a 5 wt.% Pt concentration, a typical agglomeration number is about 10% for both the Kaiser and Hydral based electrocatalysts.

Table II

X-ray Analysis of Pt Crystallite Size

Kocite Batch	Crystallite Size[a] Nominal Diameter (nm)	Agglomeration[b]	Platinum Concentration (%)
3173-173B	13.0	0.02	1.98
3173-173C	6.0	0.10	4.8
3289-25F	3.4	0.03	9.8
3380-31B	3.2	0.22	4.9
3380-31E	12.0	0.28	11.78
3380-31K	4.3	0.12	5.0
3380-104B	5.1	0.10	4.73
3380-125B	4.4	0.12	4.85

(a) mean size of crystallites over 2.5 nm diameter

(b) These results are standardized as discussed in Reference 2.

III. ELECTRODE FABRICATION AND TESTING

3.1 Electrode Fabrication

The fabrication techniques used by ERC to prepare gas diffusion fuel cell electrodes containing Kocite electrocatalysts are similar to those used to prepare ERC platinum black electrodes. The process begins with the wet blending of the electrocatalyst with a dry PTFE powder and a filler within a petroleum distillate type of medium. Considerable care must be taken during this step to prevent any Kocite particle agglomeration. After filtration of the solids, a catalyst layer is formed by a calendering operation which can be accomplished either by hand rolling or with a machine rolling mill. To improve handling during the calendering process, a decomposable filler such as ammonium bicarbonate is used. After complete vaporization of the petroleum distillate, the filler is removed by gentle heating.

The remaining steps of electrode fabrication involve pressing, mounting on a current collector, and sintering. Some variation has occurred in these steps over the duration of this program. Initially the catalyst layer was pressed at 2000 psi (high density) onto a tantalum screen current collector. More recently, a highly porous and conductive carbon fiber paper such as produced by Union Carbide Corporation has been used as a less expensive alternative to the tantalum screen current collector. The carbon fiber paper is generally wetproofed to a certain degree prior to use, by treating with FEP, a copolymer of tetrafluoroethylene and hexafluoropropylene. The pressure at which the catalyst layer is bonded to the current collector has also been varied. In the first five

month period[3] low, medium, and high density catalyst layers were prepared at 100, 500, and 2000 psi. During the second five month period a different set of bonding pressures has been made for the machine calendered electrodes. During this period these electrodes have been prepared at 2000 psi (high density), 100 psi (medium density), and by using a different filler processing step (low density).

Following the electrode bonding step, the electrode is normally sintered in a nitrogen atmosphere at 335°C for 15 minutes. Electrodes prepared from the 3380-104B and 3380-125B electrocatalysts were sintered at 345°C in order to make the sintering conditions for these Kocite electrodes identical to the present standard for Pt black electrodes.

The final thickness of the Kocite electrocatalyst layer depends on the bonding technique and pressure. For electrodes prepared during the second five month period of the contract, a 5 mg/cm^2 catalyst loading and 50 wt.% PTFE content results in catalyst layer thicknesses of 0.004", 0.006", and 0.010" for the high, medium, and low density machine calendered catalyst layers, respectively. This thickness will decrease as the PTFE content decreases.

Because of the high temperatures reached during the sintering operation, the Pt particle size was measured in a completed electrode to determine if the Pt particle size increased during electrode fabrication. In Table III, X-ray particle size results are given for both the original catalyst (125B) and two fabricated electrodes. The agglomeration is slightly larger for the electrodes in each case, but the residual crystal size is unchanged. Although the experimental situations are not identical,

Table III

Sintering of Pt in Electrodes During Fabrication

3380-125B - 5 Wt.% Pt in Catalyst
0.25 mg/cm^2 Pt Loading in Electrodes

	Pt in Catalyst	Pt in Electrode A	Pt in Electrode B
% Agglomeration[b]	13	25	27
Crystallite Size[a] (nm)	4.4	4.4	4.1

(a) mean size of crystallites over 2.5 nm diameter

(b) These results are standardized as discussed in Reference 2.

the results in Table III indicate no substantial Pt crystallite growth occurs during electrode fabrication.

3.2 Cell Testing

Details of the cell testing techniques and apparatus have been discussed in earlier reports. No changes have been made during the present period, but attempts have been made to standardize cell components. A typical cell assembly now uses pooling type graphite hardware and a high porosity Kynol matrix. At ERC, cell testing has been standardized at 149°C, the temperature ERC considers to be their baseline temperature for phosphoric acid fuel cells. Fuel cell performance at other temperatures has been measured at UOP. With one exception, to be discussed later, all cell hardware for the cells discussed in this report were of graphite and used 2" x 2" electrodes.

IV. RESULTS AND DISCUSSION

4.1 Introduction

The performance of Pt-Impregnated Kocite[R] electrodes tested at UOP and ERC is discussed in this section. Electrode characteristics and fuel cell build parameters for cells assembled at ERC are listed in Table IV. A similar listing of cell build and electrode characteristics is given in Table V for electrodes tested at UOP. At both ERC and UOP, a large number of cells were assembled. Several cells failed at ERC because of gas crossover or shorting. A larger number failed at UOP for similar reasons and cell hardware problems. Only those cell builds that performed well or were otherwise noteworthy have been listed in Tables IV and V. In most cases Kocite loadings were 5 mg/cm^2 and Pt loadings were $\sim$0.25 mg/cm^2. Teflon content, fabrication method, catalyst layer density, and variations from the normal Kocite and Pt loadings are listed in Tables IV and V. Prewetting of the Kocite electrodes by float-filling in H_3PO_4 at 140°C was continued during this reporting period although it is not clear that this is required at lower teflon contents. All cells were assembled using an ERC-fabricated 2 mg/cm^2 Pt-black electrode as the counter electrode.

The performance levels of the cells listed in Tables IV and V are summarized in Tables VI and VII. Tables VI and VII list the results obtained at ERC and UOP respectively. In these tables the full cell IR free voltages at 100 ma/cm^2 are listed when the Kocite electrode is used as an air cathode, an oxygen cathode, and a hydrogen (vs. standard air) anode. Fuel cell operation was at 149°C at ERC and varied from 140 to 180°C at UOP. Also listed is the oxygen gain for each cell. In

Table IV

Characteristics of Pt-Doped Kocite Electrodes - ERC

Cell Build Number	Kocite Batch	Nominal Kocite Loading (mg/cm^2)	Platinum Loading (mg/cm^2)	Teflon Content (Wt.%)	Fabrication Method	Relative Catalyst Layer Density	Electrode Prewetted
ERC-59	3380-31B	5.0	0.25	50	Hand-Rolled	High	1 Day
60	3380-31K	5.0	0.25	50	Hand	High	1 Day
62	3380-31E	4.25	0.50	50	Hand	High	3 Days
63	3289-25F	2.5	0.25	40	Hand	High	1 Day
64	3289-25F	2.5	0.25	40	Hand	High	1 Day
65	3380-125B	5.0	0.25	50	Hand	High	3 Days
66	3380-104B	5.0	0.25	40	Hand	High	1 Day
68	3380-125B	5.0	0.23	50	Machine	Low	1 Day
69	3380-125B	5.0	0.23	50	Machine	Low	3 Days
70	3380-125B	5.0	0.23	50	Machine	Low	2 Days
71	3380-125B	5.0	0.22	40	Machine	Low	1 Day
72	3380-125B	5.0	0.22	30	Machine	Low	1 Day
73	3380-125B	5.0	0.22	40	Machine	Low	1 Day
74	3380-125B	5.0	0.22	30	Machine	Low	1 Day
75	3380-125B	5.0	0.22	30	Machine	Medium	1 Day
76	3380-125B	5.0	0.25	50	Machine	Low	1 Day
77	3380-125B	5.0	0.25	50	Machine	Low	1 Day
78	3380-125B	5.0	0.23	40	Machine	Medium	1 Day
79	3380-125B	5.0	0.22	40	Machine	Low	3 Days
81	3380-125B	2.5	0.125	50	Machine	Low	2 Days
82	3380-125B	5.0	0.24	20	Machine	Medium	1 Day

Table V

Characteristics of Pt-Doped Kocite Electrodes - UOP

Cell Build No.	Nominal Kocite Loading (mg/cm^2)	Platinum Loading (mg/cm^2)	Teflon Content (Wt.%)	Fabrication Method	Relative Catalyst Layer Density	Electrode Prewetted
3289-25F-27[a]	5.0	0.50	30	Hand-Rolled	High	1 Day
3289-25F-28[a]	5.0	0.50	30	Hand	High	1 Day
3289-25F-30[a]	5.0	0.50	30	Hand	High	1 Day
3380-31B-16	5.0	0.25	50	Hand	High	1 Day
3380-31E-1	4.25	0.50	50	Hand	High	1 Day
3380-31K-1	5.0	0.25	50	Hand	High	1 Day
3380-104B-1	5.0	0.25	40	Hand	High	1 Day
3380-104B-2	5.0	0.25	40	Hand	High	1 Day
3380-125B-1	5.0	0.25	50	Hand	High	1 Day
3380-125B-2	5.0	0.25	50	Hand	High	1 Day
3380-125B-3	5.0	0.23	50	Machine	Low	1 Day
3380-125B-5	5.0	0.23	50	Machine	Low	1 Day
3380-125B-6	5.0	0.23	50	Machine	Low	1 Day
3380-125B-8	5.0	0.22	30	Machine	Low	1 Day
3380-125B-10	5.0	0.22	40	Machine	Low	1 Day
3380-125B-13	5.0	0.25	50	Machine	Low	1 Day
3380-125B-16	2.5	0.125	50	Machine	Low	1 Day
3380-125B-17	5.0	0.25	50	Machine	Low	1 Day
3380-125B-19	5.0	0.22	30	Machine	Low	1 Day
3380-125B-21	5.0	0.22	30	Machine	Medium	1 Day
3380-125B-24	5.0	0.22	30	Machine	Medium	1 Day
3380-125B-25	5.0	0.22	20	Machine	Medium	1 Day
3380-125B-26	5.0	0.24	20	Machine	Medium	1 Day

(a) Built with 1 in^2 tantalum current collectors, all others built with 2 in^2 graphite current collectors

Table VI

Summary of ERC Cell Performance Results at 149°C

Cell Build Number	Kocite Batch	Platinum Loading (mg/cm^2)	Teflon Content (wt.%)	Cell Voltages[a] (Volts) at 100 ma/cm^2 with Kocite Electrode at Air	O_2	H_2(vs. air)	O_2 Gain[b] mV
ERC-59	3380-31B	0.25	50	0.51[c]	0.63[c]	0.61[c]	120
ERC-60	3380-31K	0.25	50	0.61	0.69	0.63	80
ERC-62	3380-31E	0.50	50	0.43[c,d]	0.53[c,d]	0.63[c,d]	---
ERC-63	3289-25F	0.25	40	0.52	0.66	0.67	140
ERC-64	3289-25F	0.25	40	0.46	0.62	0.67	160
ERC-65	3380-125B	0.25	50	0.56	0.64	0.65	80
ERC-66	3380-104B	0.25	40	0.56	0.66	0.68	100
ERC-68	3380-125B	0.23	50	0.54	0.63	0.64	90
ERC-69	3380-125B	0.23	50	0.52[c]	0.66[c]	0.68[c]	140
ERC-70	3380-125B	0.23	50	0.56[c]	0.66[c]	0.66[c]	100
ERC-71	3380-125B	0.22	40	0.54[c]	0.66[c]	0.66[c]	120
ERC-72	3380-125B	0.22	30	0.60	0.69	0.65	90
ERC-73	3380-125B	0.22	40	0.54	0.69	0.67	150
ERC-74	3380-125B	0.22	30	0.58	0.69	0.64	110
ERC-75	3380-125B	0.22	30	0.62	0.71	0.68	90
ERC-76	3380-125B	0.25	50	0.57	0.68	0.67	110
ERC-77	3380-125B	0.25	50	0.56[c]	0.65[c]	0.66[c]	90
ERC-78	3380-125B	0.23	40	0.55	0.63	0.64	80
ERC-79	3380-125B	0.22	40	0.56[c]	0.67[c]	0.67[c]	110
ERC-81	3380-125B	0.125	50	0.54	0.64	0.67	100
ERC-82	3380-125B	0.24	20	0.57	0.67	0.64	100

[a]All cell voltages are IR free and obtained using a 2 mg/cm^2 Pt-black counter electrode.

[b]Voltage difference between air and oxygen performance at 100 ma/cm^2.

[c]Estimated IR correction of 60 mV because the resistance was not directly measured.

[d]at 60 ma/cm^2, 30 mV IR correction.

Table VII

Summary of UOP Cell Performance Results

Cell Build Number	Platinum Loading (mg/cm^2)	Teflon Content (%)	Cell Voltages[b] (Volts) · 100 ma/cm^2 with Kocite electrode on Air	O_2	H_2(vs. air)	O_2 gain[c] mV	Temperature °C
3289-25F-27[a]	0.50	30			0.704		180
3289-25F-28[a]	0.50	30			0.747		180
3289-25F-30[a]	0.50	30	0.500	0.730	0.654	230	180
3380-31B-16	0.25	50	0.537	0.626	0.590	90	140
3380-31E-1	0.50	50	0.537	0.645		108	140
3380-31K-1	0.25	50	0.519	0.632		100	140
3380-104B-1	0.25	40	0.535	0.640		105	140
			0.550	0.630		80	160
			0.565	0.655		91	180
3380-104B-2	0.25	40	0.532	0.628		96	140
			0.532	0.633		101	160
			0.554	0.653		99	180
3380-125B-1	0.25	50	0.547	0.622		75	140
			0.570	0.648		78	160
			0.542	0.651		109	180
3380-125B-2	0.25	50	0.531	0.626		95	140
3380-125B-3	0.23	50	0.553	0.656		103	140
			0.573	0.673		100	160
			0.562	0.655		93	180
3380-125B-5	0.23	50	0.532	0.633		101	140
			0.573	0.677		104	160
			0.602	0.690		88	180
3380-125B-6	0.23	50	0.556	0.671		115	140
			0.573	0.685		112	160
3380-125B-8	0.22	30	0.572	0.680		108	140
			0.588	0.689		101	160
			0.611	0.695		84	180
3380-125B-10	0.22	40	0.485				140
3380-125B-13	0.25	50	0.471	0.602		131	140
			0.605	0.693		88	180
3380-125B-16	0.125	50	0.543	0.650		107	140
			0.566	0.670		104	160
			0.581	0.667		86	180

Table VII (Continued)

Summary of UOP Cell Performance Results (Continued)

Cell Build Number	Platinum Loading (mg/cm^2)	Teflon Content (%)	Cell Voltages(b) (Volts) 100 ma/cm^2 with Kocite electrode on Air	O_2	H_2 (vs. air)	O_2 gain(c) mV	Temperature °C
3380-125B-17	0.25	50	0.522	0.642		120	140
3380-125B-19	0.22	30	0.454	0.587	0.636	133	140
3380-125B-21	0.22	30	0.570	0.675		105	160
3380-125B-24	0.22	30	0.570	0.670		100	160
3380-125B-25	0.22	30	0.590	0.680		90	160
3380-125B-26	0.24	20	0.600	0.705		105	160

(a) Built using 1 in^2 tantalum current collectors, all others built with 2 in^2 graphite current collectors.

(b) All cell voltages are IR free and obtained using a 2 mg/cm^2 Pt black counter electrode

(c) Voltage difference between air and oxygen performance at 100 ma/cm^2

general, comparable performance levels were obtained at ERC and UOP with these electrodes.

4.2 Performance of Individual Kocite^R Batches

4.2.1 Kocite Batch 3289-25F

The performance of Kocite electrodes fabricated from Kocite batch 3289-25F has been discussed in detail in the previous Interim Report.(3) It is with these electrodes that the best performance to date had been obtained for Kocite electrodes used as air or oxygen cathodes. Typically, Kocite electrodes have not performed well as anodes. However, one Kocite electrode from this batch performed stably at 180°C for 500 hours. The cell with this electrode was first run at 140°C for 100 hours and then at 180°C for an extra 500 hours. Scanning electron microscopy microprobe measurements were made when the cell was dismantled to insure that the anode had the correct Pt concentration and had not been switched with a Pt-black electrode having four times the Pt-loading.

The full cell polarization curve obtained when this electrode was used as a hydrogen (vs. air) anode at 180°C is shown in Figure 1. The polarization curve was obtained after 500 hours of operation at 180°C. This cell is the only one discussed in the current report which used tantalum hardware and an electrode size of 1" X 1". The voltage output of this cell at 100 ma/cm^2 and at both 140 and 180°C is shown in Figure 2. Both terminal and IR corrected voltages are shown. While the terminal voltage decreased somewhat after 500 hours operations at

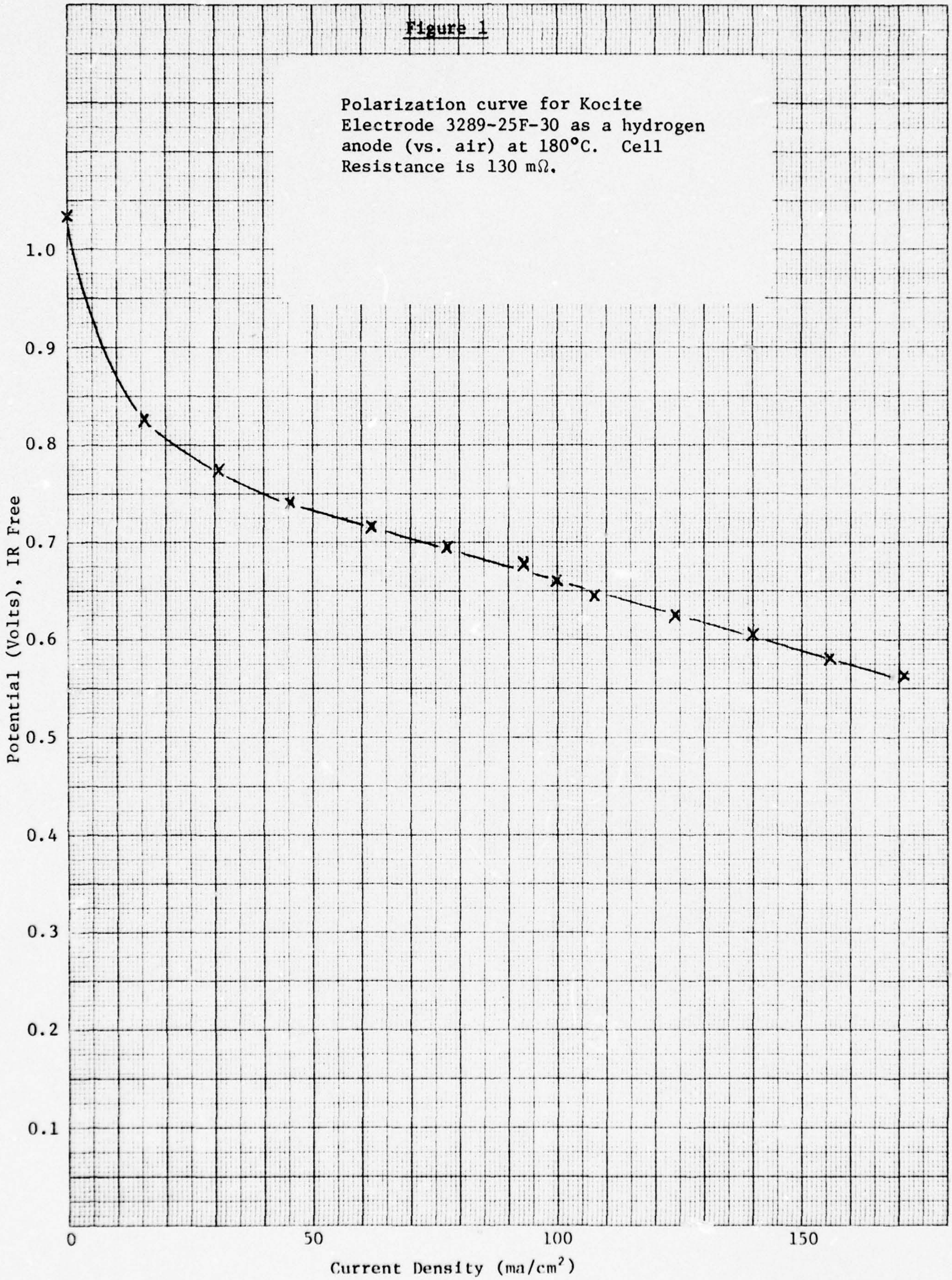
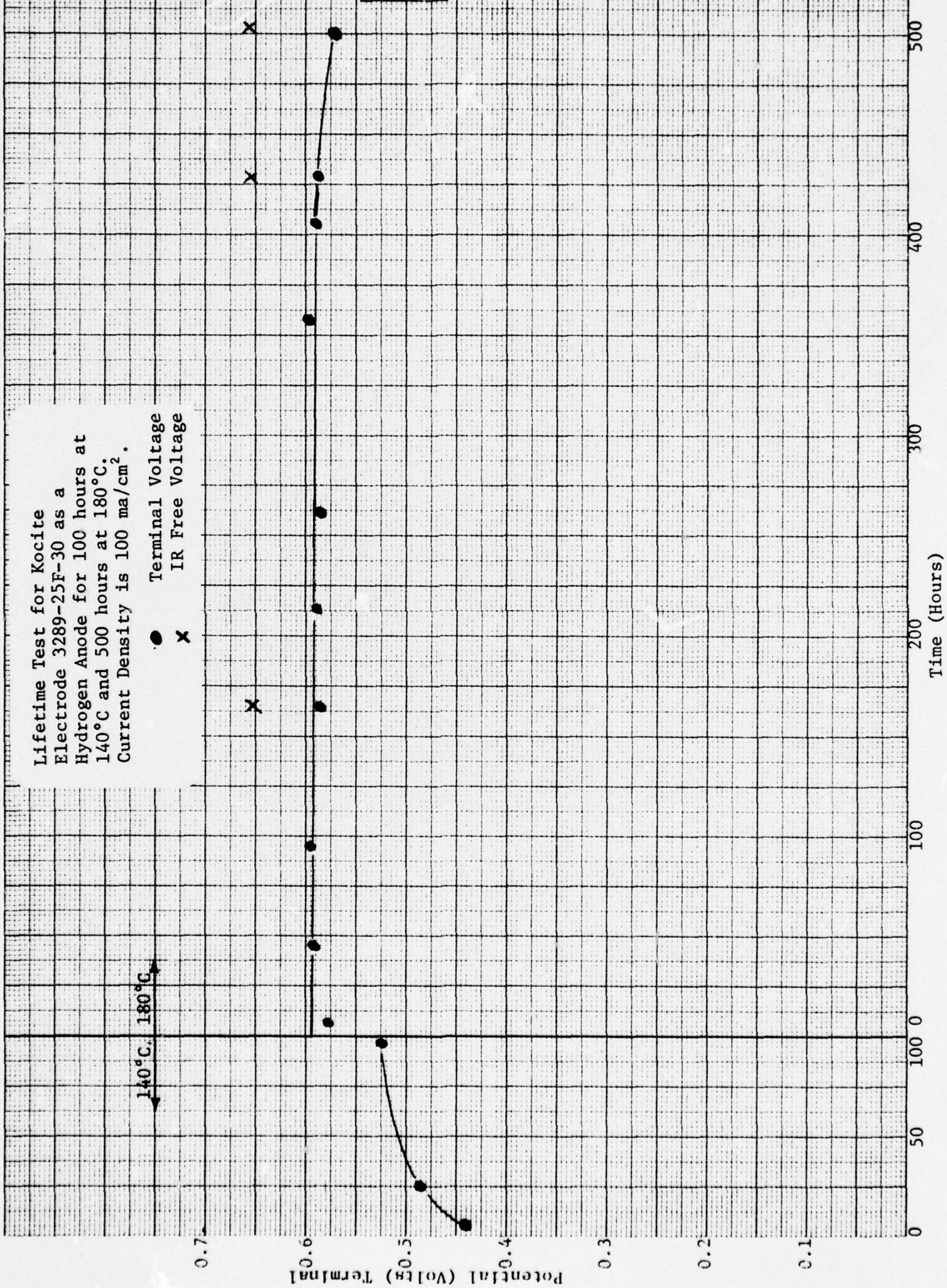


Figure 1

Polarization curve for Kocite Electrode 3289-25F-30 as a hydrogen anode (vs. air) at 180°C. Cell Resistance is 130 mΩ.

Figure 2


Lifetime Test for Kocite Electrode 3289-25F-30 as a Hydrogen Anode for 100 hours at 140°C and 500 hours at 180°C. Current Density is 100 ma/cm^2.
● Terminal Voltage
× IR Free Voltage
140°C
180°C
Time (Hours)
0
50
100
0
100
200
300
400
500
Potential (Volts) Terminal
0.7
0.6
0.5
0.4
0.3
0.2
0.1

180°C, the IR corrected voltage remained unchanged. These results indicate no electrocatalyst deterioration occured during this period when this Kocite electrode was run as an anode at 180°C for 500 hours. The increase in the cell resistance at 180°C temperature is not surprising and probably results from corrosion of the cell components or some matrix dryout. While the data shown in Figures 1 and 2 are not typical of Kocite electrodes used as anodes, they show that Kocite electrodes are capable of use as anodes and that a fuel cell can be constructed using only Kocite electrodes.

4.2.2 Kocite Batch 3380-31B

Pt loading - 0.25 mg/cm^2

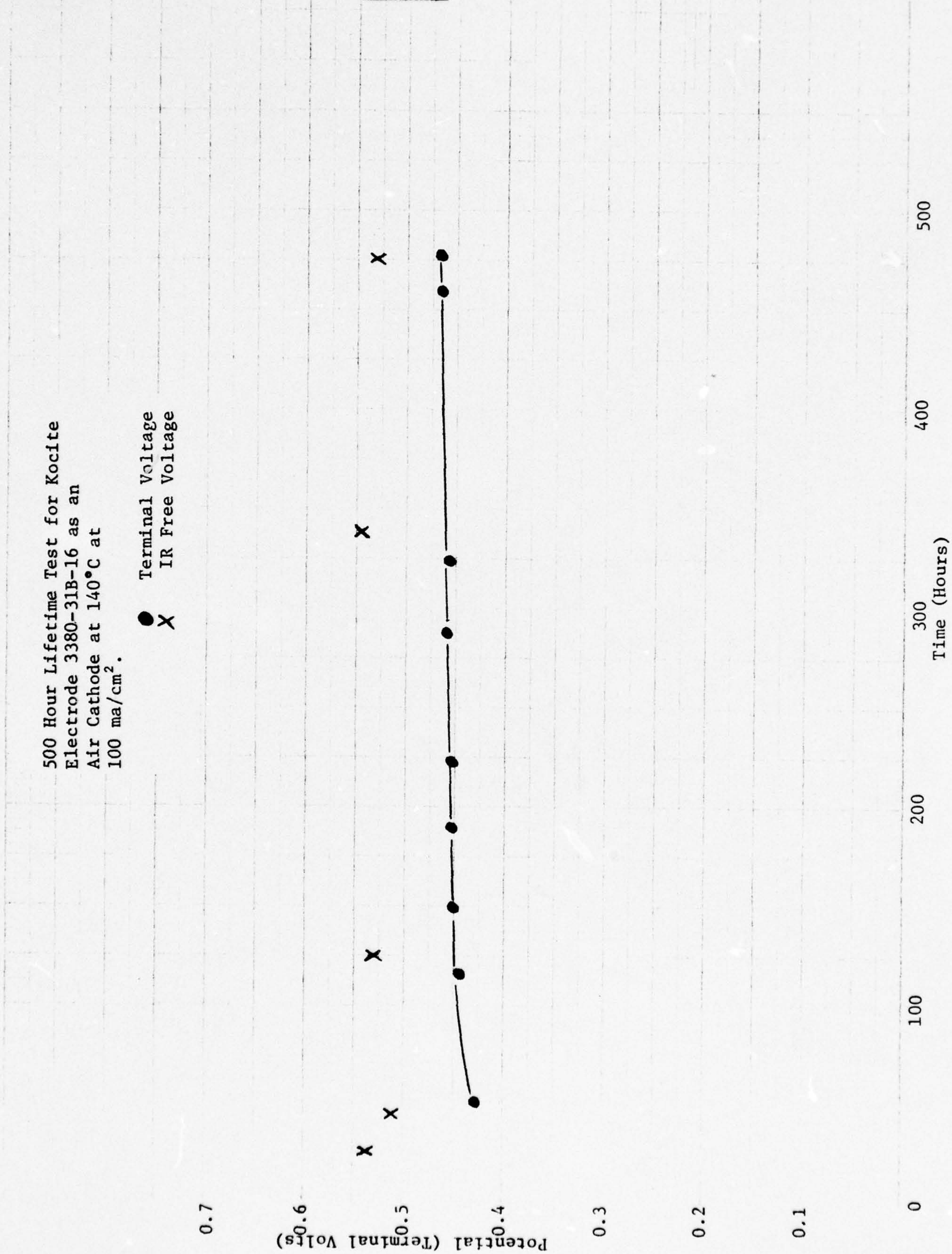
The performance of electrodes fabricated from Kocite Batch 3380-31B was discussed in detail in the previous Interim Report.(3) For completeness, a 500 hour lifetime test is included which shows the Hydral based electrocatalyst performs stably as an air cathode at 140°C. In Figure 3,both the terminal and IR free voltages are shown for a cell using this electrocatalyst. The terminal voltage increases with time while the IR free voltage remains stable. Since the catalyst layer of this electrode has a 50 wt.% teflon content, and is difficult to wet with H_3PO_4, the slow increase of performance with time probably results from a gradual wetting of the catalyst layer.

4.2.3 Kocite Batch 3380-31E

Pt loading - 0.50 mg/cm^2

The best results with electrodes fabricated from this batch are shown in Figure 4, where polarization curves at 140°C are shown with the Kocite electrode used as an air cathode and an oxygen cathode.

Figure 3


500 Hour Lifetime Test for Kocite Electrode 3380-31B-16 as an Air Cathode at 140°C at 100 ma/cm^2.
● Terminal Voltage
X IR Free Voltage
Potential (Terminal Volts)
0.7
0.6
0.5
0.4
0.3
0.2
0.1
0
100
200
300
400
500
Time (Hours)

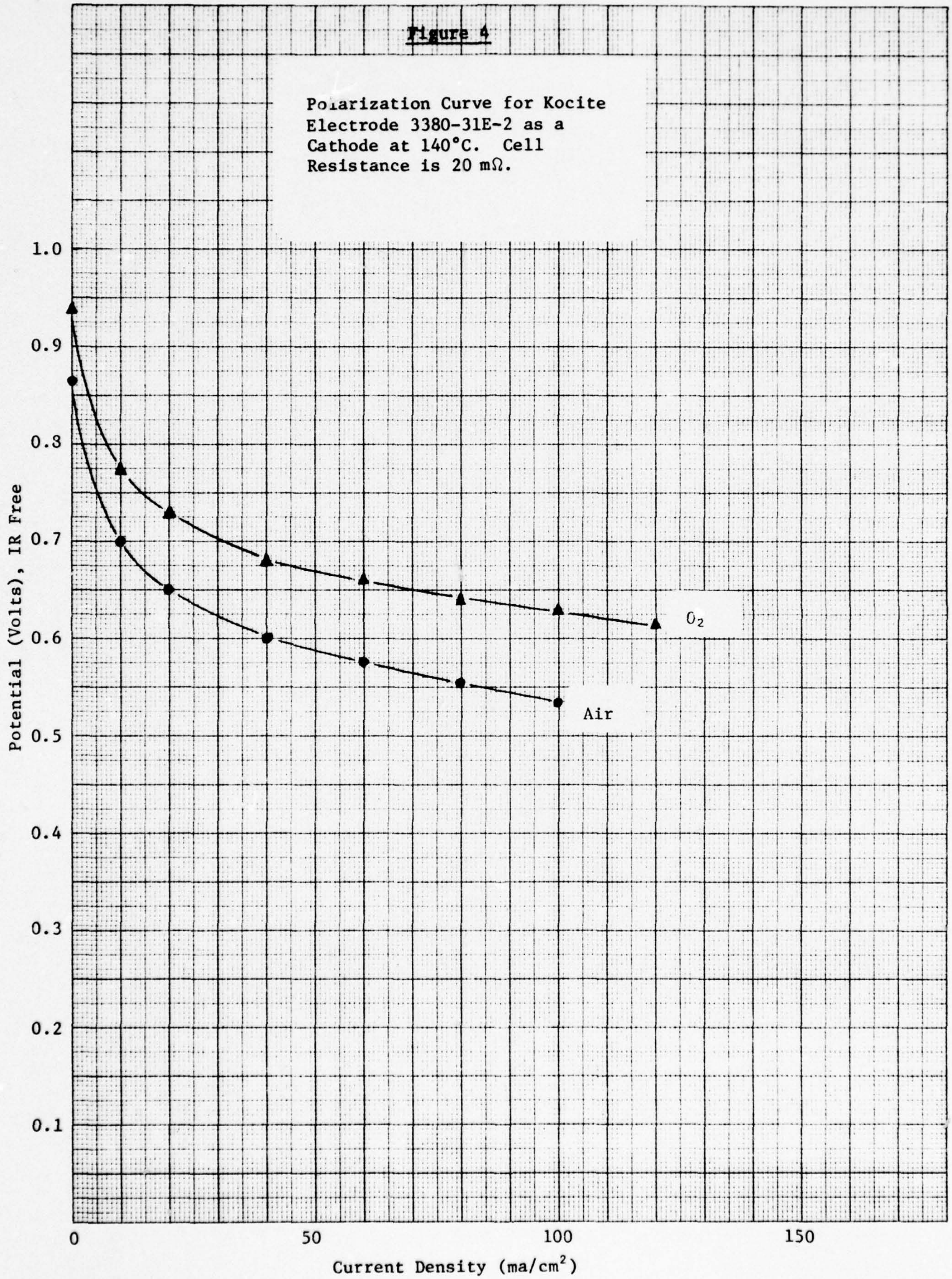


Figure 4
Polarization Curve for Kocite Electrode 3380-31E-2 as a Cathode at 140°C. Cell Resistance is 20 mΩ.
1.0
0.9
0.8
0.7
0.6
0.5
0.4
0.3
0.2
0.1
Potential (Volts), IR Free
O2
Air
0
50
100
150
Current Density (ma/cm2)

Considering the high Pt loading, electrodes from this batch did not perform well either at UOP or ERC even though the oxygen gain was less than 100 mV (normally consistent with high performance levels). Further testing of electrodes from this batch was discontinued.

4.2.4 Kocite Batch 3380-31K

Pt loading - 0.25 mg/cm^2

The results obtained with electrodes fabricated from Kocite batch 3380-31K were very encouraging. At this Pt loading a terminal voltage of 0.56 volts at 100 ma/cm^2 and 149°C was obtained by ERC. IR-free fuel cell voltages of 0.61 on air and 0.69 on oxygen were achieved at this temperature and current. This performance is 60 mV better than previously obtained with a 0.25 mg/cm^2 Pt-loaded air cathode and 30 mV better than previously obtained with an oxygen cathode. Not only do these results show a higher performance level than previously achieved, but also indicate that higher performance levels can be achieved with a Hydral based electrocatalyst than have been attained with a Kaiser based electrocatalyst.

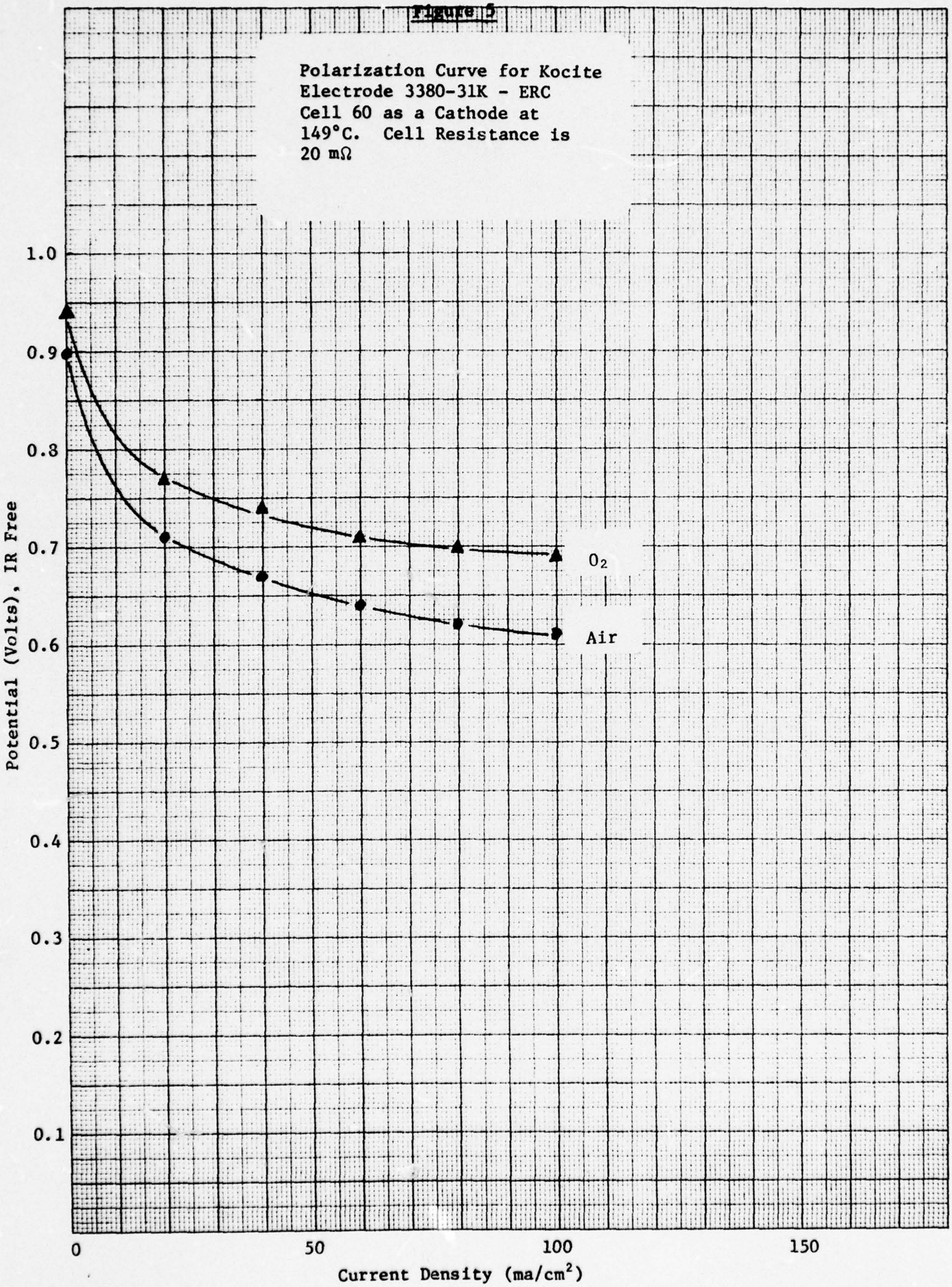
In Figure 5, full cell polarization curves are shown for the cell having the best performance using an electrode from this Kocite batch. The polarization curves in this figure were obtained using the Kocite electrode as both an air and oxygen cathode at 149°C. At 100 ma/cm^2, the oxygen gain figure for this cell is just 80 mV.

4.2.5 Kocite Batch 3380-104B

Pt loading - 0.25 mg/cm^2

This electrocatalyst batch is the only one tested to date

Figure 5


Polarization Curve for Kocite Electrode 3380-31K - ERC Cell 60 as a Cathode at 149°C. Cell Resistance is 20 mΩ
1.0
0.9
0.8
0.7
0.6
0.5
0.4
0.3
0.2
0.1
Potential (Volts), IR Free
O2
Air
0
50
100
150
Current Density (ma/cm2)

using the $\sim 1.2 \times 10^3$ nm diameter particle-sized Kaiser Medium Alumina as the electrocatalyst base. The best performance obtained with an electrode from this Kocite batch is shown in Figure 6, where full cell polarization curves are shown when the Kocite electrode is used as an air and oxygen cathode at 149°C. With the exception of the best performance obtained using an electrode from Kocite Batch 3380-31K, the performance levels shown in Figure 6 exceed those of any 0.25 mg/cm^2 Pt-loaded electrode tested previously. The results of an 800 hour air cathode lifetime test at 149°C and 100 ma/cm^2 are shown in Figure 7. In this figure the terminal voltage of the cell is shown when the Kocite electrode is tested as an air or oxygen cathode. The air cathode performance is stable during this lifetime test while the oxygen performance declined by about 10 mV.

Although the performance levels obtained with electrodes from batch 3380-104B are close to those obtained with batch 3380-31K and 3380-125B (discussed in the next section), further testing of electrodes from this and related batches using the small-sized ground Kaiser alumina base have been temporarily discontinued. After the electrode structure has been optimized for the Hydral based electo-catalysts, some additional testing will be performed with Kaiser based electrocatalysts.

4.2.6 Kocite Batch 3380-125B

Pt loadings - 0.125 mg/cm^2 to 0.25 mg/cm^2

This electrocatalyst batch is the first larger batch ($\sim$200 gms)

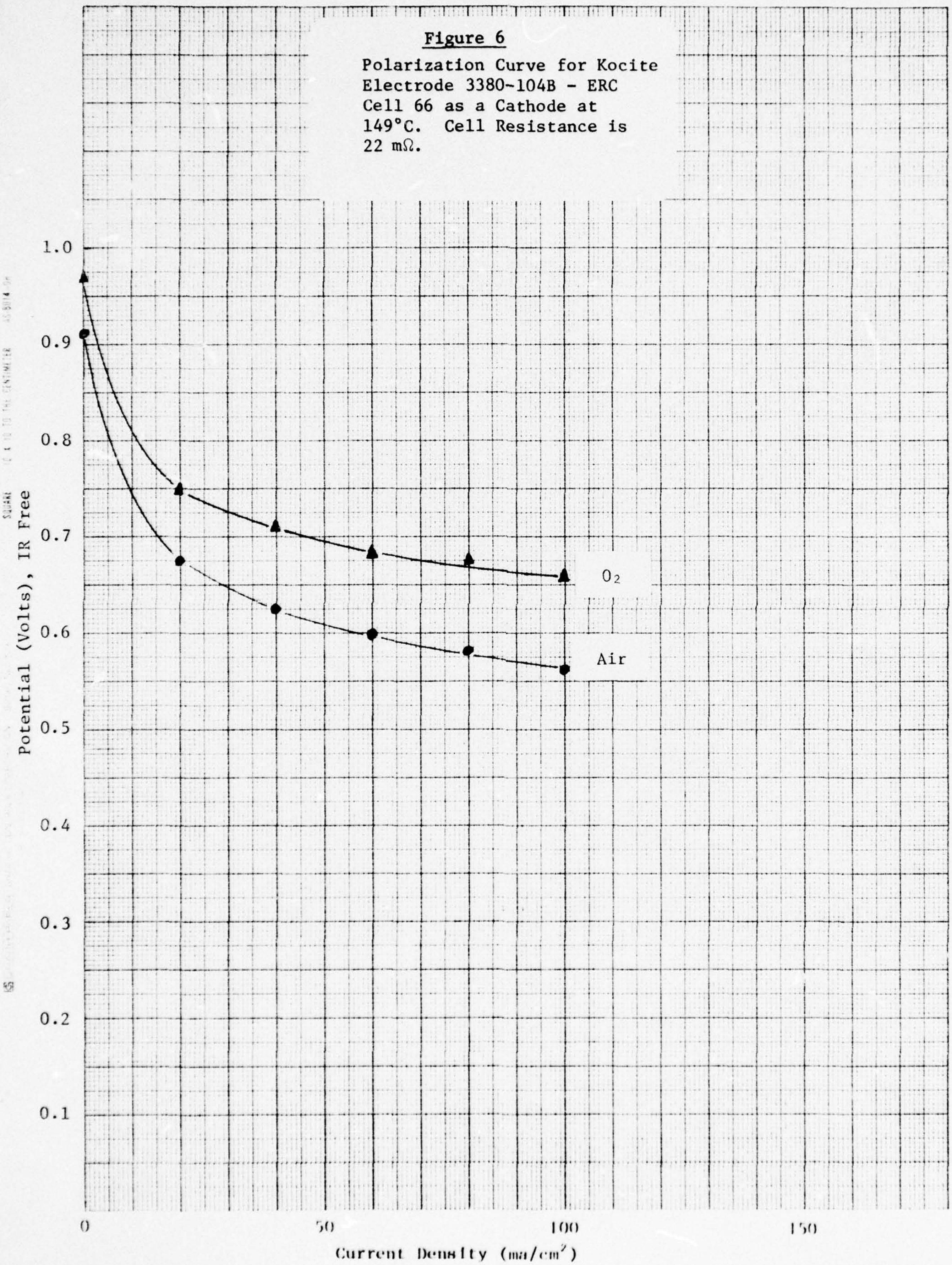
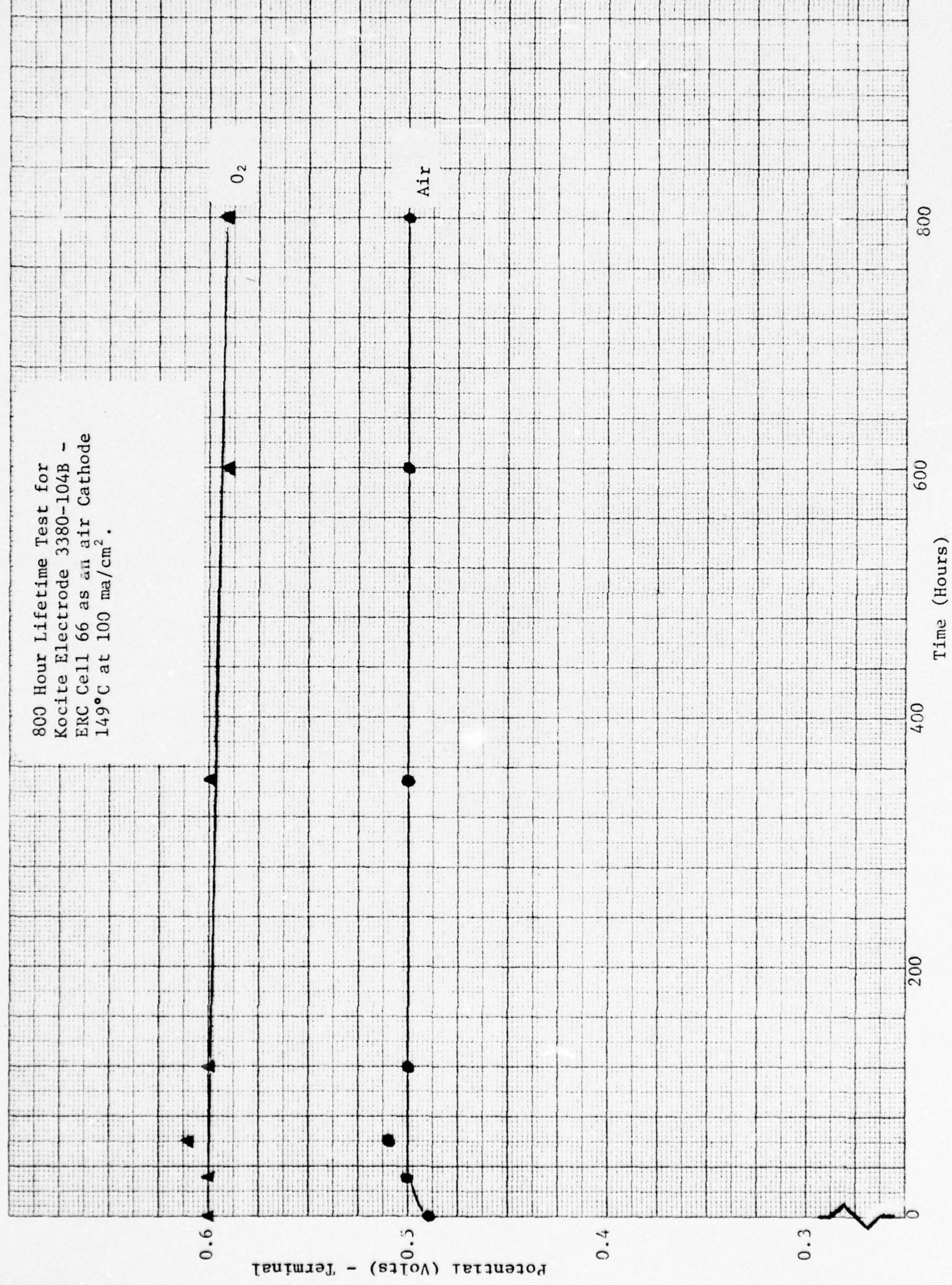


Figure 6
Polarization Curve for Kocite Electrode 3380-104B - ERC Cell 66 as a Cathode at 149°C. Cell Resistance is 22 mΩ.
1.0
0.9
0.8
0.7
0.6
0.5
0.4
0.3
0.2
0.1
Potential (Volts), IR Free
O2
Air
0
50
100
150
Current Density (ma/cm²)

Figure 7


800 Hour Lifetime Test for
Kocite Electrode 3380-104B -
ERC Cell 66 as an air Cathode
149°C at 100 ma/cm².
O2
Air
Potential (Volts) - Terminal
0.6
0.5
0.4
0.3
Time (Hours)
0
200
400
600
800

to be fabricated into electrodes. The majority of the cell testing during this reporting period has involved either hand or machine calendered electrodes from this batch. The best performance levels obtained to date are with an electrode from batch 3380-125B. At a Pt loading of 0.22 mg/cm^2, using the Kocite electrode as an air cathode at 149°C and 100 ma/cm^2, a cell terminal voltage of 0.57 volts was achieved. The IR free performance levels of this cell on air and oxygen are 0.62 and 0.71 volts, respectively, giving an oxygen gain figure of 90 mV under these operating conditions. Polarization curves obtained with this cell using the Kocite electrode as an air and oxygen cathode are shown in Figure 8.

Good performance levels have also been achieved with other cells using electrodes fabricated from Kocite batch 3380-125B. Full cell polarization curves obtained for a cell operated at 160°C are shown in Figure 9. The oxygen gain figure for this cell is 80 mV at 100 ma/cm^2.

Due to a lack of cell hardware, few lifetime tests have been performed with electrodes from this batch. The results of an 800 hour air cathode lifetime test at ERC are shown in Figure 10. The terminal voltage of the cell at various times during the test was measured with no evidence of any deterioration of cell performance when the Kocite electrode was used either as an air or oxygen cathode.

One lifetime test was performed with a cell using a 0.125 mg/cm^2 Pt-loaded Kocite electrode having a catalyst layer of half the

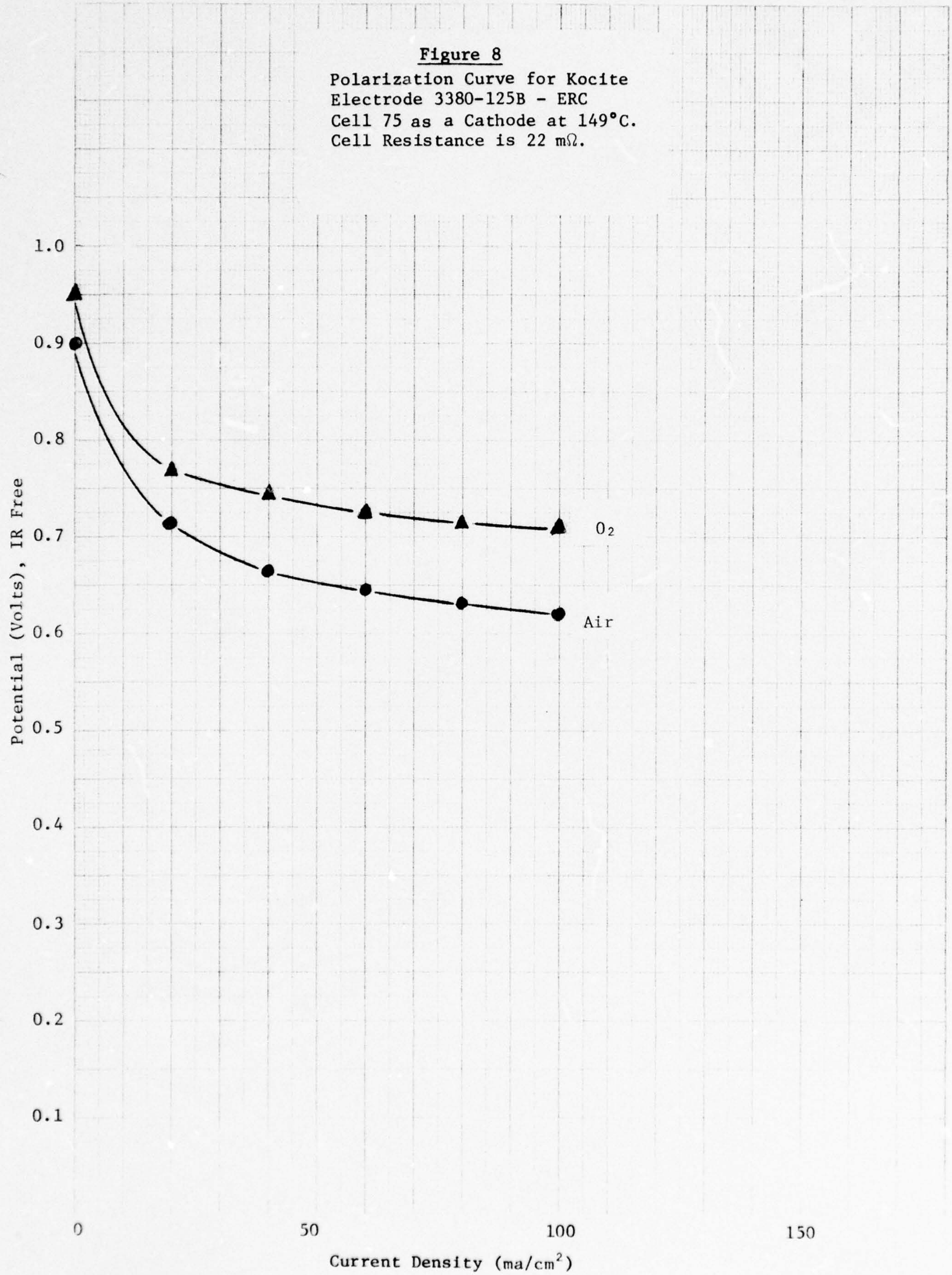


Figure 8
Polarization Curve for Kocite
Electrode 3380-125B - ERC
Cell 75 as a Cathode at 149°C.
Cell Resistance is 22 mΩ.
1.0
0.9
0.8
0.7
0.6
0.5
0.4
0.3
0.2
0.1
Potential (Volts), IR Free
0
50
100
150
Current Density (ma/cm²)
O2
Air

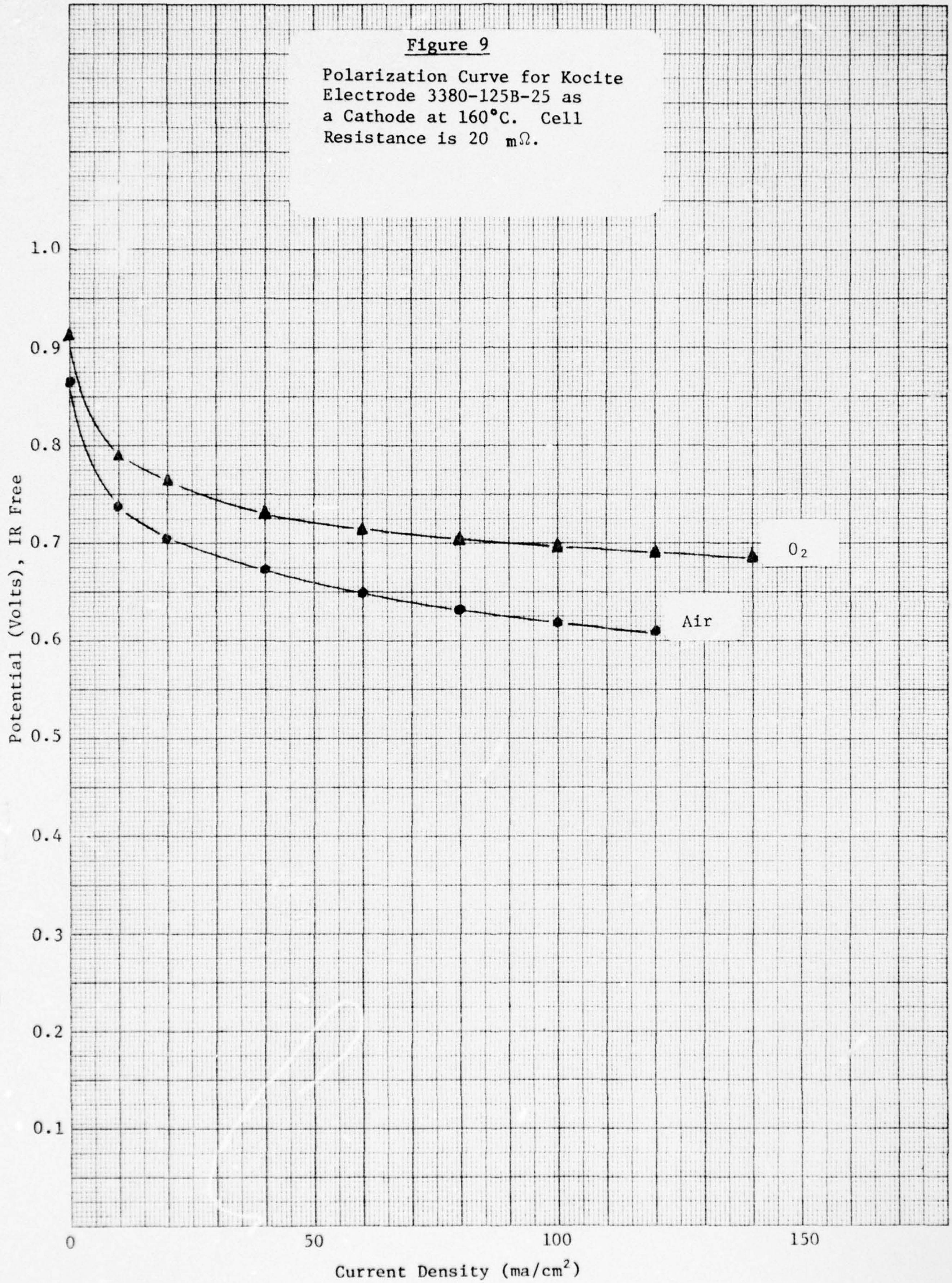
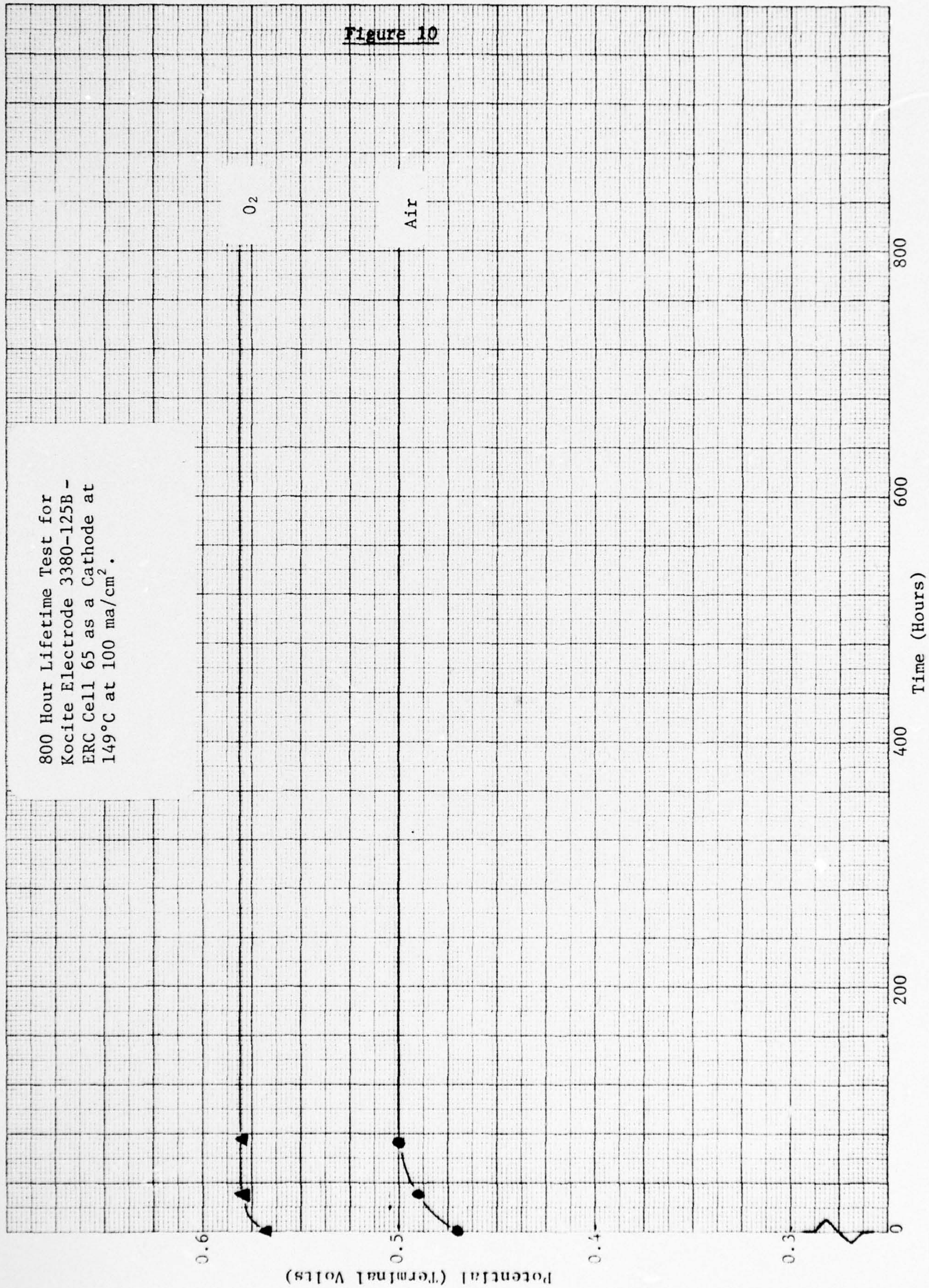


Figure 9
Polarization Curve for Kocite Electrode 3380-125B-25 as a Cathode at 160°C. Cell Resistance is 20 mΩ.
Potential (Volts), IR Free
1.0
0.9
0.8
0.7
0.6
0.5
0.4
0.3
0.2
0.1
0
50
100
150
Current Density (ma/cm²)
O₂
Air
SQUARE 10 X 10 TO THE CENTIMETER

Figure 10


800 Hour Lifetime Test for
Kocite Electrode 3380-125B -
ERC Cell 65 as a Cathode at
149°C at 100 ma/cm².
O2
Air
Potential (Terminal Volts)
0.6
0.5
0.4
0.3
Time (Hours)
0
200
400
600
800

customary thickness. The results of this test are shown in Figure 11. The Kocite electrode was used first as an air cathode at 140°C and 100 ma/cm^2 for about 340 hours. Then the temperature was raised to 160°C. At both temperatures the cell performed without deterioration, although, for this cell the cell resistance was higher at the higher temperature. This test indicates that no lifetime problems are to be expected using Kocite electrodes with a thinner catalyst layer.

A large number of electrodes have been fabricated from this batch and the structural properties of these electrodes have been varied systematically. Some testing of these electrodes has been completed but many cells are still in test. The results of the tests completed to date are summarized in the next section.

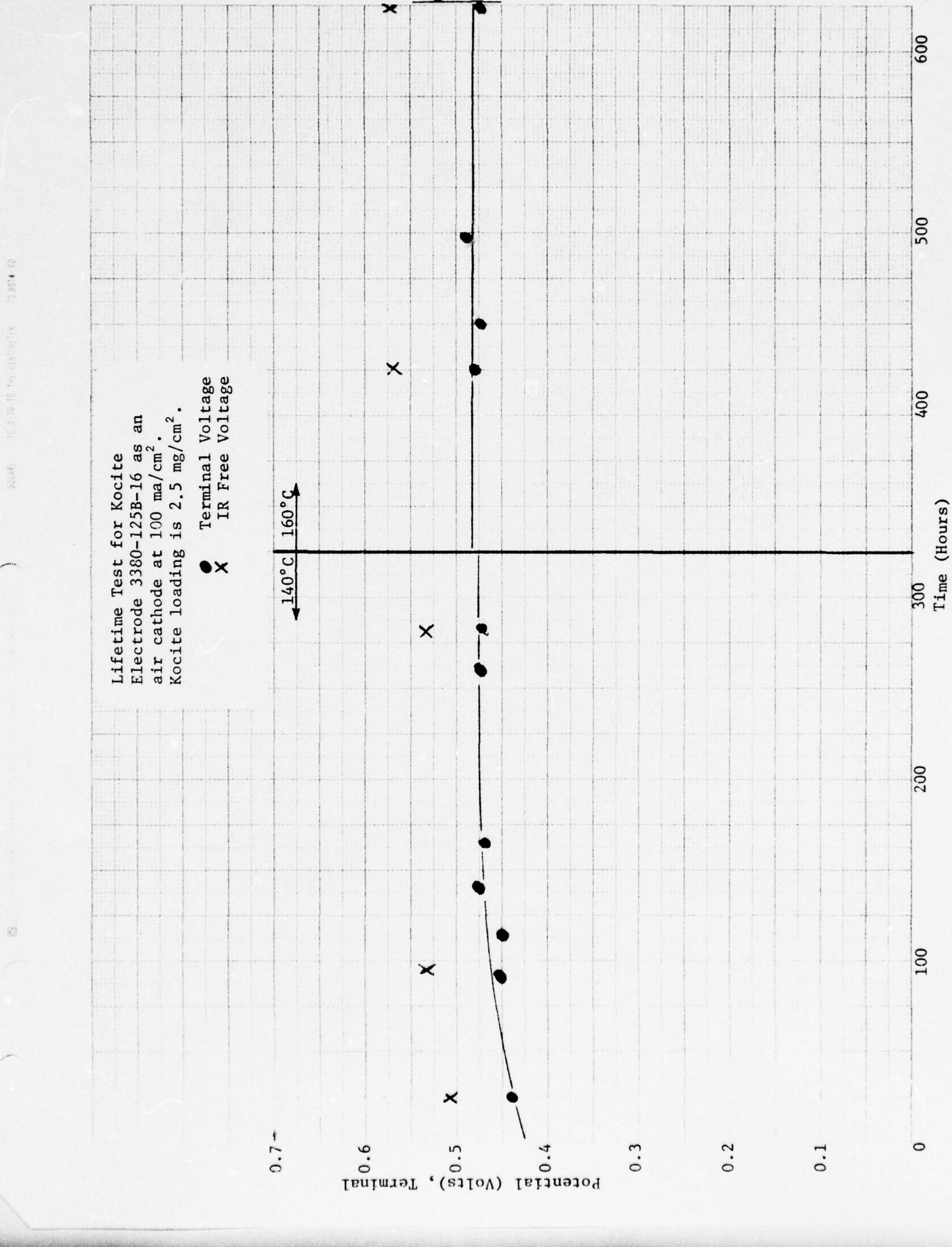
4.3 The Effects of Electrode Structure on Kocite^R Electrode Performance

4.3.1 Introduction

Using machine calendered electrodes from Kocite batch 3380-125B to insure reproducibility of electrode fabrication, the structural properties of Kocite electrodes are being varied systematically. The effects of three electrode parameters are currently under investigation. These parameters are:

- Teflon content of catalyst layer
- Density of catalyst layer
- Catalyst layer thickness

Figure 11


Lifetime Test for Kocite
Electrode 3380-125B-16 as an
air cathode at 100 ma/cm^2.
Kocite loading is 2.5 mg/cm^2.
Terminal Voltage
IR Free Voltage
140°C
160°C
Potential (Volts), Terminal
0.7
0.6
0.5
0.4
0.3
0.2
0.1
0
100
200
300
400
500
600
Time (Hours)

While the systematic variation of these parameters has not been completed, sufficient test data have been accumulated to indicate the type of electrode structure which should give the best performance for Kocite electrodes.

During electrode fabrication, problems were encountered relating to the tendency of Kocite electrocatalysts to agglomerate into $(50\text{-}100) \times 10^3$ nm size clumps. These agglomerates can easily be broken apart during the blending step of electrode fabrication, but considerable care is required to ensure that Kocite agglomeration is minimized. To reduce this problem, ERC has added a dry-blending step preceding to the normal wet-blending step. This additional step definitely reduced the agglomeration problem. Scanning electron microscope analysis of the surface of the electrodes as received by UOP indicates some agglomeration remains and varies with electrode batch. As a result, the air cathode performance and perhaps the wetting properties of the electrode are probably affected to some degree. In general, the machine calendered electrodes tested to date have a slightly higher oxygen gain figure ($\sim$90-100mV) than the best hand rolled electrodes ($\sim$75-90mV). It is believed that longer blending times will reduce this problem.

4.3.2 Variation of Cell Performance with Teflon Content

Kocite electrodes have been fabricated from Kocite Batch 3380-125B with catalyst layer teflon contents of 20, 30, 40, and 50 wt.%. At the higher teflon contents, wetting of the Kocite electrode is difficult so that over a hundred hours is normally required for the cell to approach maximum performance. The dependence of maximum cell performance on

teflon content is shown in Figure 12a for a series of low density electrodes with a 5 mg/cm^2 Kocite loading. These results were obtained at ERC. The cells were operated with the Kocite electrode as an air and oxygen cathode at 149°C and 100 ma/cm^2. Both the air and oxygen performance levels are shown in Figure 12a. Several features of the data merit discussion. Clearly, the best air cathode performance levels are obtained at lower teflon contents indicating the optimum teflon content is in the range of 20-40 wt.%. Oxygen cathode performance appears to peak at 30 or 40 wt.% teflon, however, the electrode with the 20 wt.% teflon content catalyst layer may not have achieved its maximum performance level by the time of this writing. The substantial scatter in the performance of the cells with Kocite electrodes containing 50 wt.% teflon catalyst layers is probably due to both the problem of Kocite agglomeration encountered in the earliest machine calendered electrodes and the difficulty of uniformly wetting Kocite electrodes with high teflon content catalyst layers.

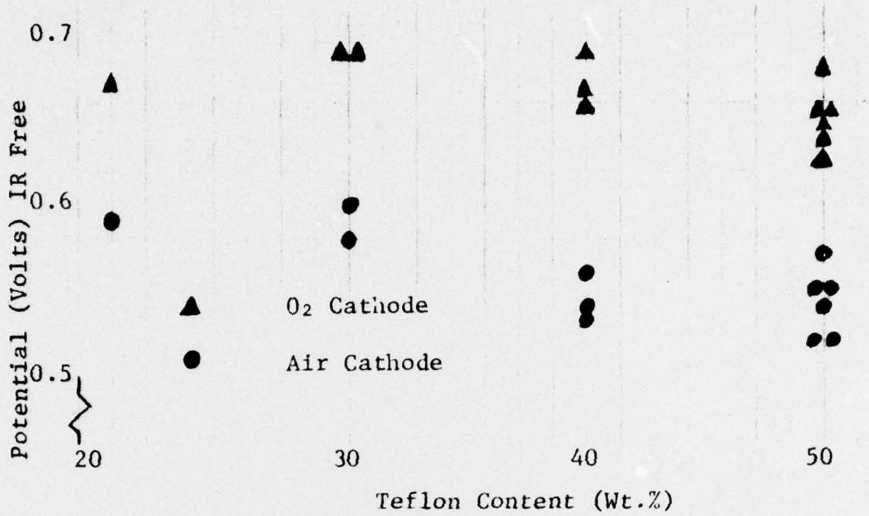
The dependence of cell performance on the Kocite electrode teflon content at 160 and 180°C is shown in Figure 12b. Results are shown for both air and oxygen cathode performance. At higher temperatures the performance levels appear to be less dependent on teflon content although electrodes having a catalyst layer with the lower teflon content give a slightly better performance than those with higher teflon content (the performance for the electrode with a 20% teflon content catalyst layer may not have reached maximum). The weaker dependence on teflon content at higher temperature may result from improved wetting for electrodes with

Figure 12

12 a

Effect of Teflon Content on Cell Performance
ERC Results.

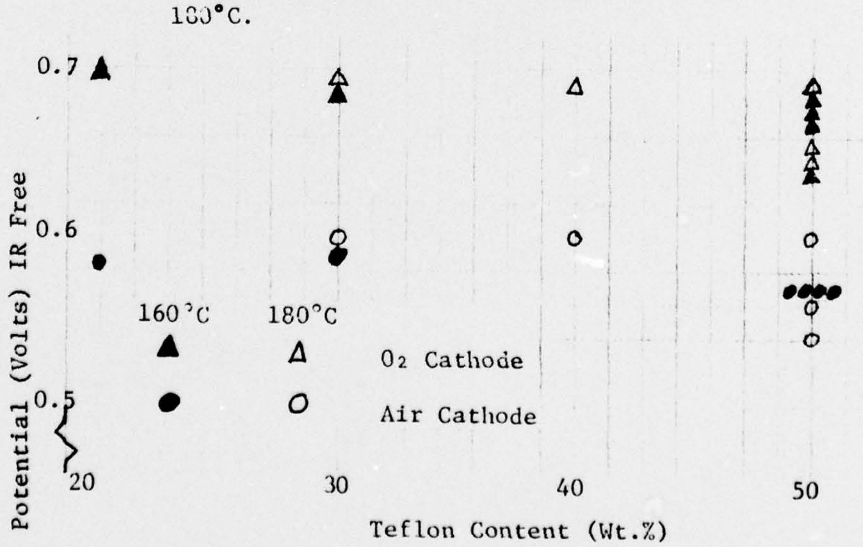
Voltage, IR Free, is plotted at 100 ma/cm^2 for machine rolled electrodes at 149°C.



12 b

Effect of Teflon Content on Cell Performance
UOP Results

Voltage, IR Free, at 100 ma/cm^2 is plotted for machine rolled electrodes at 160°C and 180°C.



the higher teflon content. Testing of the effect of teflon content on electrode performance is continuing.

4.3.3 Variation of Cell Performance with Catalyst Layer Density

The dependence of electrode performance on catalyst layer density is being determined for electrodes fabricated from Kocite batch 3380-125B. To date, results have been obtained on machine calendered electrodes with low or medium density catalyst layers. Several sets of electrodes have been tested where Kocite loading, teflon content, etc., have been held constant and the density varied as described in section 3.1. The results obtained to date in these tests are summarized in Table VIII. The best performance data (see Figure 8) have been obtained using a medium density electrode with a 30 wt.% teflon content. However, a comparison of the results obtained for the different sets of electrodes listed in Table VIII shows no clear difference between electrodes with low and medium density catalyst layers. Testing of electrodes with different density catalyst layers is continuing.

4.3.4 Variation of Cell Performance with Catalyst Layer Thickness

Using Kocite batch 3380-125B, electrodes with different Kocite loadings and hence catalyst-layer thickness have been fabricated. In order to determine the importance of catalyst-layer thickness several sets of cells can be compared where the catalyst layer of the Kocite electrode has the same teflon content and density.

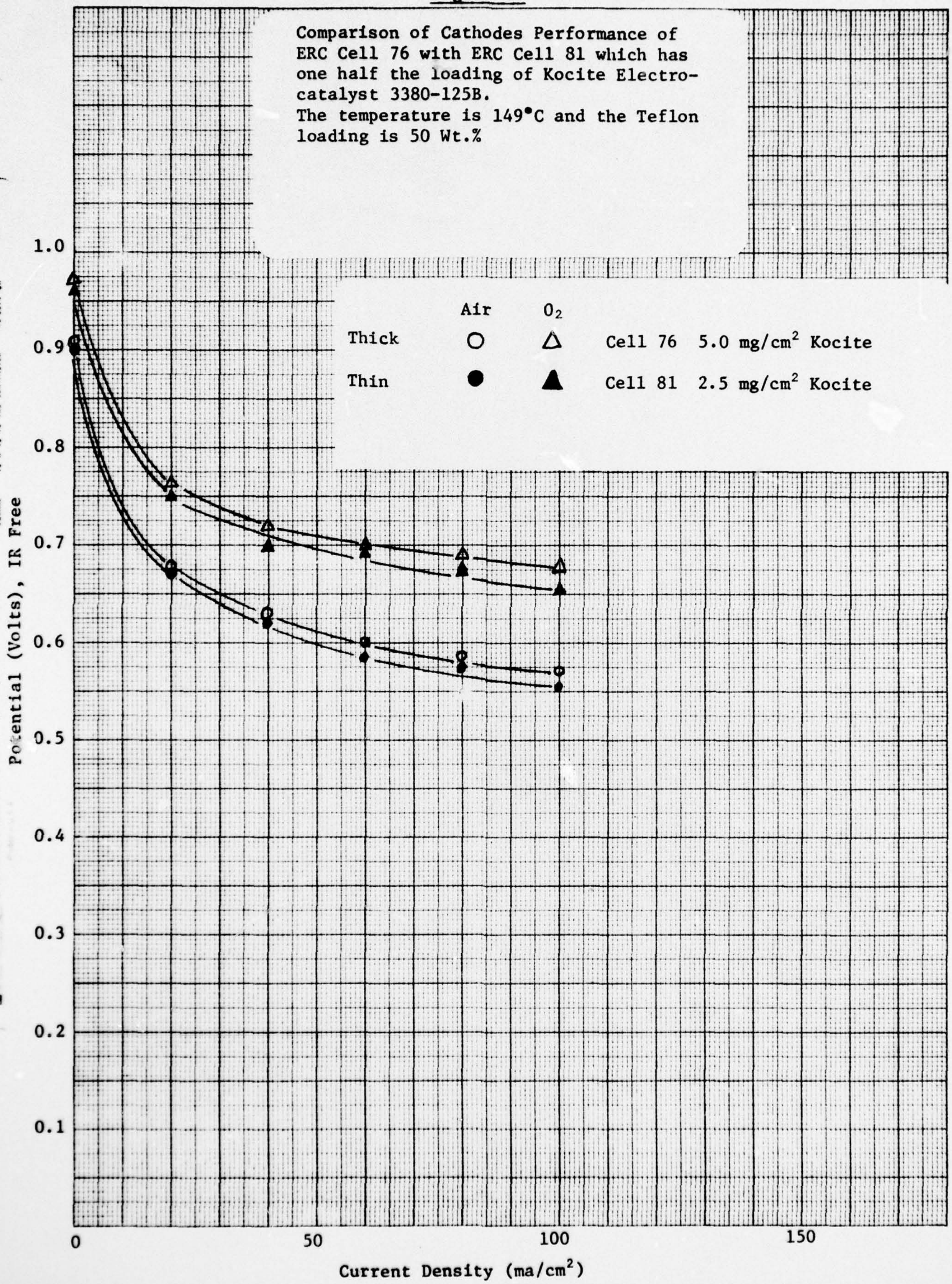
A comparison of two cells, each having Kocite electrodes with low density catalyst layers containing 50 wt.% teflon, is shown in Figure 13. The air cathode and oxygen cathode full cell polarization curves indicate

Table VIII

Catalyst Density Effects on Average Cell Performance

Cell Conditions	Density	Cell Voltage IR Free 100 ma/cm^2 Air	O_2
UOP Cells with 125B Electrodes	Low	0.59	0.69
0.22 mg/cm^2 Pt, 30 Wt.% Teflon, 160°C	Medium	0.58	0.68
ERC Cells with 125B Electrodes	Low	0.59	0.69
0.22 mg/cm^2 Pt, 30 Wt.% Teflon, 149°C	Medium	0.62	0.71
ERC Cells with 125B Electrodes	Low	0.55	0.67
0.22 mg/cm^2 Pt, 40 Wt.% Teflon, 149°C	Medium	0.55	0.63

Figure 13

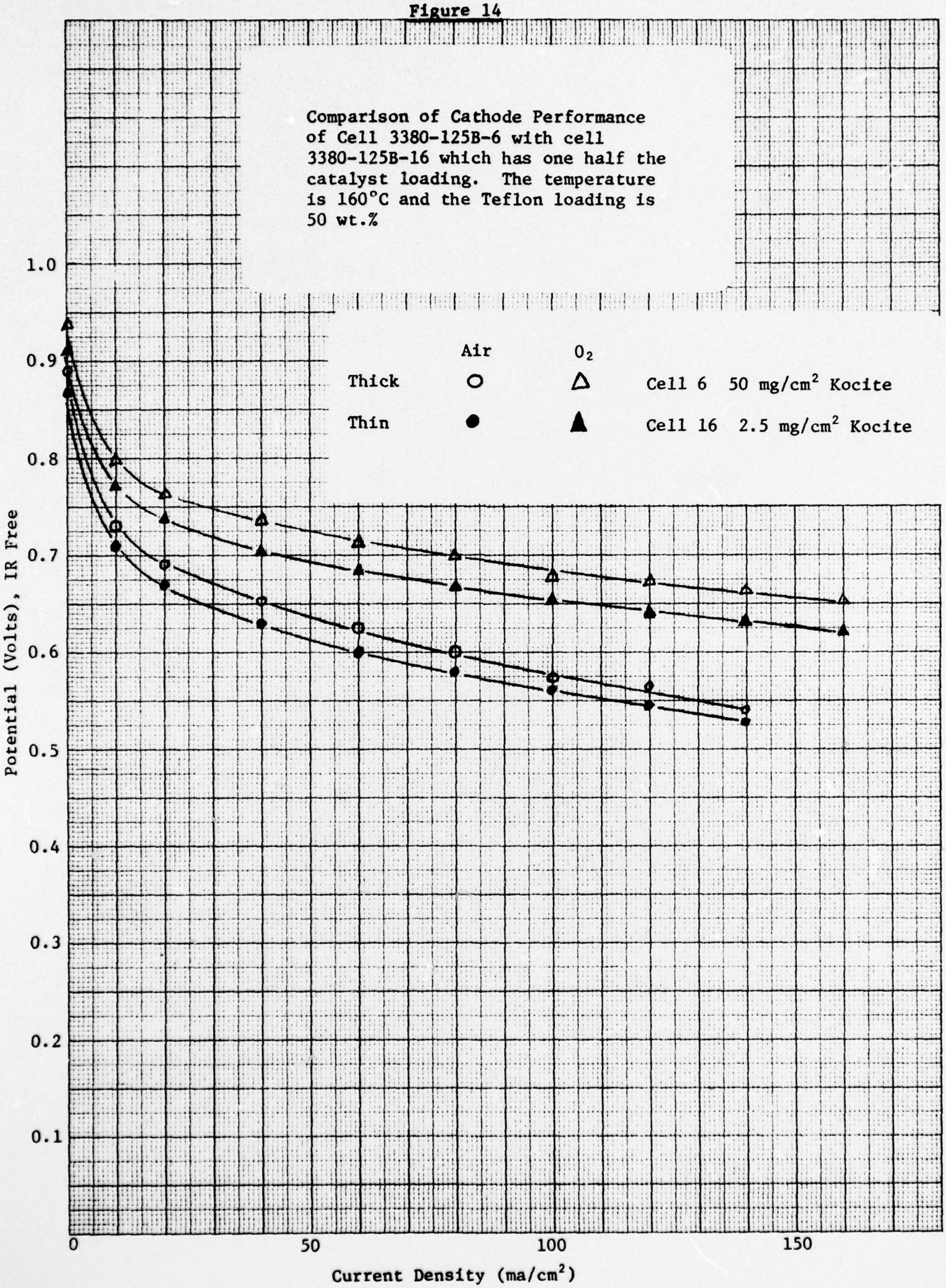

Comparison of Cathodes Performance of ERC Cell 76 with ERC Cell 81 which has one half the loading of Kocite Electro-catalyst 3380-125B.
The temperature is 149°C and the Teflon loading is 50 Wt.%
Air
O2
Thick
Cell 76 5.0 mg/cm2 Kocite
Thin
Cell 81 2.5 mg/cm2 Kocite
1.0
0.9
0.8
0.7
0.6
0.5
0.4
0.3
0.2
0.1
Potential (Volts), IR Free
0
50
100
150
Current Density (ma/cm2)

very similar performance although the catalyst layer thickness and Pt loading of one cell is half that of the other. Improved performance may be possible with the use of thinner electrodes, if the voltage of the lower Pt-loaded cell at 50 ma/cm^2 is taken as an indication of the expected performance of the thinner electrode at 100 ma/cm^2 (if its Pt-loading is doubled). (As shown earlier[3], electrode performance is roughly proportional to Pt-loading.) From the data shown in Figure 13, such a comparison would suggest a 30 mV improvement might be obtained for an air cathode and a 20 mV improvement for an oxygen cathode.

A similar comparison is made in Figure 14 for two cells operated at 160°C with electrodes having low density catalyst layers of 50 wt.% teflon content. In this case the higher Pt-loaded electrode has a catalyst layer thickness 1.84 times that of the thinner catalyst layer electrode. A comparison similar to the one above can be made from Figure 14 of the voltage of the thinner electrode at 50 ma/cm^2 to the voltage of the thicker electrode at 100 ma/cm^2. This provides an estimate of the performance of a thinner electrode having the same platinum loading. In this case, the air cathode improvement would also be 30 mV while the oxygen cathode improvement would be about 5mV.

These results indicate a noticeable improvement in Kocite electrode performance may be achieved with the use of thinner catalyst layers. Higher Pt concentration Kocite electrocatalysts have been prepared (up to 20 wt.% Pt) and will be tested to determine if the expected improvement in performance can be attained.

Figure 14


Comparison of Cathode Performance of Cell 3380-125B-6 with cell 3380-125B-16 which has one half the catalyst loading. The temperature is 160°C and the Teflon loading is 50 wt.%
Air
O_2
Thick
Thin
Cell 6 50 mg/cm² Kocite
Cell 16 2.5 mg/cm² Kocite
Potential (Volts), IR Free
1.0
0.9
0.8
0.7
0.6
0.5
0.4
0.3
0.2
0.1
0
50
100
150
Current Density (ma/cm²)
SQUARE 10 X 10 TO THE CENTIMETER 46 0804-04

4.4 Summary of Results

Over the second five months of this contract period, several important results have been obtained which are listed below.

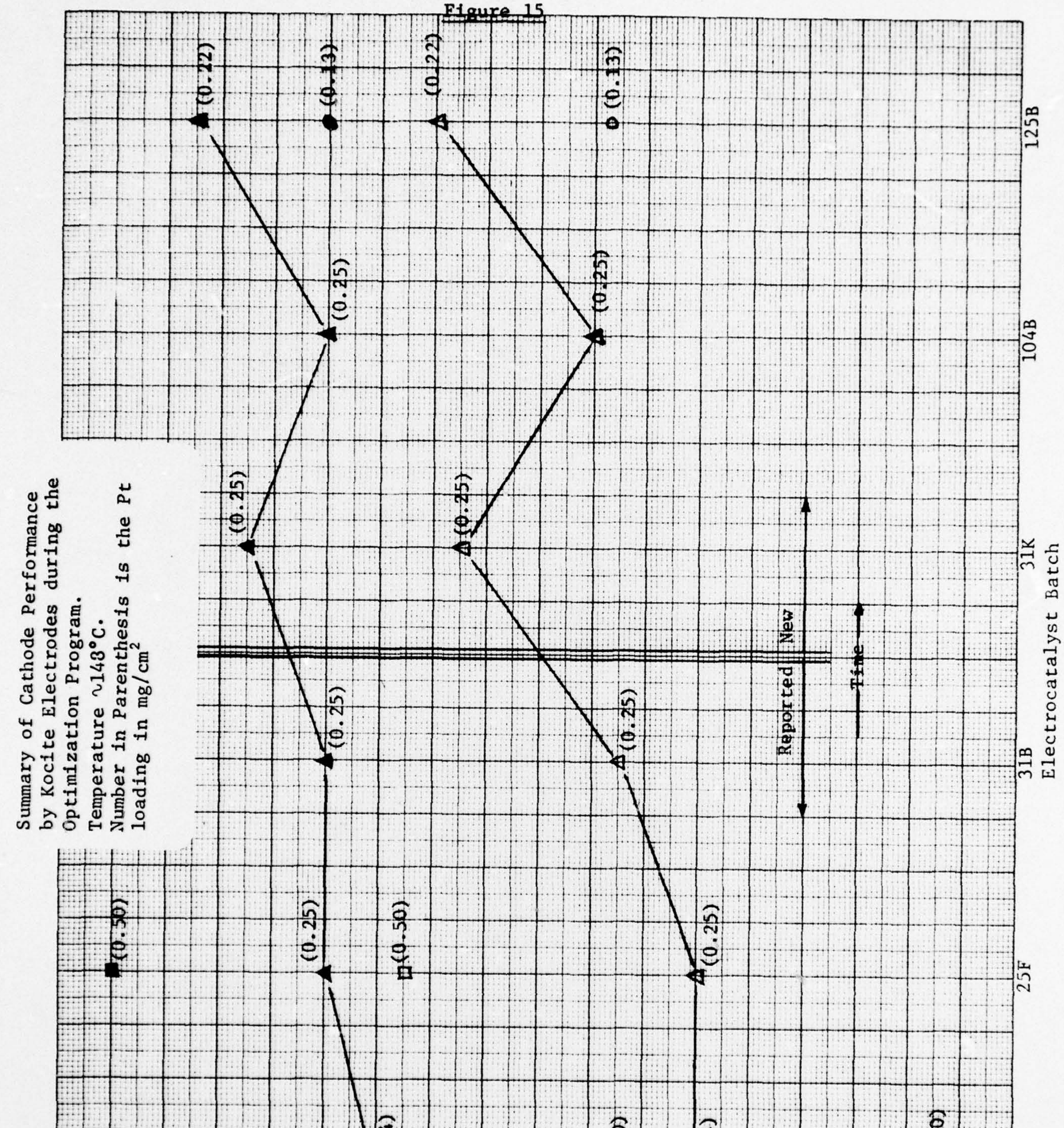
(A) Kocite Electrocatalyst Production

Several large ($\geq$ 200 gms) batches of Kocite electrocatalysts have been prepared and sent to ERC for incorporation into electrodes. Batches of this size allow larger electrode batches to be prepared by ERC using machine calendering techniques to assure electrode uniformity and reproducibility. Several of these batches have used a commercially available alumina base, Hydral 705.

(B) Electrode Performance

The improvement in electrode performance obtained during the current contract is documented in Figure 15. Starting with electrodes from Kocite batch 3173-173C, the improvement in the performance achieved with $\sim$0.25 mg/cm^2 Pt-loaded electrodes used as air or oxygen cathodes is shown by the solid lines of this figure. The current best air cathode performance of a Kocite electrode from Kocite batch 3380-125B is 100 mV better than that of the best air cathode performance of a 3173-173C batch electrode. Also shown in this figure is the best performance achieved with a 0.50 mg/cm^2 Pt-loaded 3189-25F batch electrode and the performance of the lower loaded ($\sim$0.1 mg/cm^2 Pt) batch 3173B and 3380-125B electrodes. Over this period the air cathode, oxygen cathode and oxygen gain figure have been improved considerably. From the results obtained from the electrode structure optimization tests, it is expected

Figure 15


Summary of Cathode Performance by Kocite Electrodes during the Optimization Program.
Temperature ∿143°C.
Number in Parenthesis is the Pt loading in mg/cm²
Voltage at 100 ma/cm² - IR Free
0.70
0.60
0.50
0.40
Electrocatalyst Batch
173 B&C
25F
31B
31K
104B
125B
Reported
New
Time
(0.24)
(0.10)
(0.24)
(0.10)
(0.50)
(0.25)
(0.50)
(0.25)
(0.25)
(0.25)
(0.25)
(0.25)
(0.25)
(0.25)
(0.22)
(0.13)
(0.22)
(0.13)

substantial improvements in electrode performance will be made before the end of the contract period.

For the first time stable anode performance has been achieved with a Kocite electrode. At 180°C the IR free performance of this electrode showed no deterioration after 500 hours operation.

(C) Electrode Structure Optimization

Efforts are in progress to optimize the teflon content, density, and thickness of the Kocite catalyst layer. The results obtained to date indicate the following:

1. Teflon Content - Better electrode performance, particularly below 160°C, is obtained when the catalyst layer teflon content is below 40 wt.% teflon. At this time, 30 wt.% teflon is probably close to the optimum teflon content.
2. Catalyst Layer Density - Although the best performance results to date have been obtained with a medium density catalyst layer, the data are insufficient to determine the optimum density.
3. Catalyst Layer Thickness - The use of electrodes with thinner catalyst layers (∿0.003" or less) is expected to give some improvement in electrode performance. It is estimated that this improvement may be as large as 30 mV for the air cathode.

In these areas the test data are not yet complete. Work is continuing with the goal of determining the optimum electrode structure by the end of this contract period.

V. CONCLUSIONS

Based on improved results obtained in the last five months, several conclusions concerning low cost fuel cell electrodes using Kocite materials can be stated.

A. Kocite electrodes can be used as stable cathodes and anodes at 180°C.

B. At Pt loadings between 0.25 and 0.50 mg/cm^2, terminal voltage performance levels of 0.60 volts at 100 ma/cm^2 should be obtainable with Kocite electrodes used as air cathodes between 160 and 180°C. At 100 ma/cm^2, the best terminal voltage results obtained to date are 0.57 volts at 149°C with a 0.22 mg/cm^2 Pt loading, and 0.59 volts at 165°C with a 0.5 mg/cm^2 Pt loading.

C. Progress has been made in the optimization of the Kocite electrode structure. Results to date indicate that teflon content of the catalyst layer should be near 30 wt.% and the Kocite loading reduced to 2.5 mg/cm^2 or lower.

VI. FUTURE WORK

During the remainder of the current contract period, the efforts of ERC and UOP will be concentrated in the following areas.

A. Kocite Production

Several large batches will be prepared using pyropolymer precursors other than benzene to attain a more hydrophilic catalyst. Pyridine, decane, and furan appear to be likely choices.

B. Pt-Impregnation

Several large Kocite electrocatalyst batches will be prepared with Pt concentrations up to 20 wt.% to allow fabrication of thinner catalyst layers with 0.25 and 0.5 mg/cm^2 loadings.

C. Anode Poisoning

Either co-impregnation techniques or additives will be employed in an attempt to reduce the poisoning of the anode.

D. Electrode Optimization

The thickness, density, teflon content, etc., of the catalyst layer of the Kocite electrodes will continue to be varied in order to determine the optimum structure. This will continue to be the main part of the program and will require extensive electrode testing. Efforts will be directed toward thinner electrode catalyst layers as this appears to offer the best possibility for improved electrode performance.

E. X-ray Sintering Studies

The degree of Pt particle growth during cell operation will be determined by comparing the degree of Pt agglomeration in the as-

received electrode with that after 500 hours of operation. The results of these X-ray measurements will be compared against those of Pt-impregnated carbon black electrodes.

VII. REFERENCES

1. L. B. Welsh, G. L. Hervert, D. H. Spielberg, K. J. Youtsey, "Carbonaceous Catalysts for H_3PO_4 Fuel Cells", MERDC Contract DAAK02-75-C-0011, Interim Technical Report, March, 1975.

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3. L. B. Welsh, R. W. Leyerle, G. L. Hervert, "Optimization of Pt-Doped Kocite[R] Electrodes in H_3PO_4 Fuel Cells", MERADCOM Contract DAAG53-76-C-0014, Interim Technical Report, March, 1976.